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Low voltage water electrolysis: Decoupling hydrogen production using bioelectrochemical system

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ABSTRACT

Decoupling water electrolysis using mediator is an interesting way to produce pure hydrogen. The present work validates the proof of concept of decoupled electrolyser associated with a bioelectrochemical system (MFC-DES) through a redox flow mediator (potassium hexacyanoferrate (KHCF)). Low voltage (1 V) hydrogen production was achieved with a current density up to 25 mA cm⁻². Regeneration of the mediator was performed by glucose fed microbial fuel cells. The oxidation rate of KHCF in the electrolyser is, at least, an order of magnitude higher than the reduction rate in MFC cascade fed system. MFC-DES is thus a promising set up as it desynchronizes limited microbial rate and hydrogen production, generate value from wastewater and reduce energetic cost of water electrolysis.

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Introduction

The growing need for energy by the human society and depletion of conventional energy sources demands renewable, safe, low-cost and omnipresent energy sources. Bioelectrochemical system (BES) have gained interest over the last decade to convert biomass wastes into bioelectricity and chemicals [1]. Indeed, it's possible to take advantage of

interactions between bacteria and electrodes to run electrochemical reaction [2]. In conjunction with this strategy, power-to-gas and in particular power-to-hydrogen shall be part of our future (renewable) energy mix [3]. At present, most H₂ produced has a fossil origin and mankind strives to develop renewable H₂ production, e.g. by water electrolysis (J. Mergel, 2014) based on the electrochemical reaction of water splitting:



List of acronyms: BES, Bioelectrochemical system; COD, chemical oxygen demand; DES, decoupling electrolysis system; ECPB, electron-coupled proton buffer; HCF, hexacyanoferrate; KHCF, potassium hexacyanoferrate; MEC, microbial electrolysis cell; MES, microbial electrosynthesis; MFC, microbial fuel cell; OER, oxygen evolution reaction; RFB, redox flow battery; RHE, reversible hydrogen electrode.

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The minimum applied voltage needed to decompose water at standard conditions, is equal to 1.229 V at 25 °C and it is also called the standard reversible cell potential, E^0 given by:

$$E^0 = \frac{-\Delta G^0}{2F} \quad (2)$$

with ΔG^0 the standard Gibbs free energy of water (237.14 kJ mol⁻¹) and F the Faraday constant the gas constant (96485 C mol⁻¹). In addition, the water electrolysis process introduces several barriers that shall be broken through. Thus, three types of barriers were distinguished, i) the transport of species such as ions and electrons (resistance of interconnection), resistance of ionic transport through the electrolyte and resistance due to the gas bubbles (O₂ and H₂), ii) the reaction kinetics iii) the minimal thermodynamic voltage E^0 .

The first category of resistance or overvoltage (related to the transport of species) is mainly addressed by cell design optimization in order to minimize these effects. To decrease the magnitude of resistances due to electrochemical kinetics, it is necessary to develop new catalysts materials specifically for oxygen evolution reaction (OER). Nowadays OER catalysts as Ruthenium oxide (RuO₂) and Iridium oxide (IrO₂) catalysts are among the most active reported to date, but due to their low elemental abundance and high cost, the use of these materials limits large spread of water electrolysis [4]. To make possible the dream of clean and usable energy, hydrogen production shall be provided from numerous strategy of renewable energy.

An alternative approach to the conventional water electrolysis process is to divide water splitting into two steps, whereby the electrons and protons generated are taken up during oxygen production thanks to an electron-coupled proton buffer (ECPB), instead of being used directly to produce H₂ [5]. Numerous advantages are highlighted by using this strategy, but the most important one is that H₂ production can be evolved at elevated pressures and be performed at a rate dissociated to the rate of the first step [6]. Therefore, using a redox mediator to split the conventional water electrolysis process into separate H₂ and O₂ production steps, overcomes the gas-mixing issues and increases the use of renewable energy. In the strategy of decoupling electrolysis or redox flow battery (RFB [7] [8]), mediators are managed independently to cope with the low energy intensity and the intermittency of renewable energy sources. Therefore it is possible to take advantage of the research in the domain of RFB [9] and of the recent development of ECPB to propose a new strategy of BES applications.

Microbial Fuel Cell (MFC) has firstly been proposed as an innovative solution to convert bio-degradable substrate directly into electricity while treating wastewater [10,11]. However, even if high coulombic efficiency were reported at lab scale, several limitations were encountered while scaling up [12] and using complex effluent [13]. The most challenging one is the air-cathode due to the still unsolved equation between high oxygen overpotential (specially with low anodic current density) and material costs [14]. In spite of design optimization due to extensive investigation in the past decade, it likely appears that direct electricity recovery in air-cathode MFC from wastewater to power devices or national

grid is still unrealistic [15] and hardly competitive with well-controlled and widely diffused anaerobic digestion technology. In a second time, Microbial Electrolysis Cell (MEC) has been presented as a competitive application to produce hydrogen from biowaste reducing energy costs compared to water electrolysis (0.4–1 V vs. 1.2–1.6 V) [16]. Nevertheless, major bottlenecks still include pH gradient in reactors with membrane, hydrogen scavenger losses and gas purity in free membrane systems [17]. Coupled MFC and MEC system have been successfully proposed to produce hydrogen without energy input [18]. However, this system is synchronized to air-cathode MFC performance, which limits voltage and current density leading to a small conversion rate. Lastly, microbial electrosynthesis is most likely to be part of the promising BES application to produce added values chemicals and/or store carbon dioxide [19]. Cathode reactions are mainly targeted leading to several challenges such as extracellular electron transfer comprehension in cathode (for non-abiotic system), rate synchronization with anode reaction and economic viability (chemicals products prices and electricity cost vs. conventional industrial production cost) [20]. Even if BES is full of promising perspectives, major system limitations need to be overcome and niche applications have yet to be found.

In the present work, we combine a cascade-fed MFC set-up with an electrolyser using the commonly used redox flow mediator potassium hexacyanoferrate (KHCF) (II) and (III), noted respectively HCF(II) and HCF(III) (Fig. 1). Systems are running independently at different current density. In the biologic reactor, organics acid oxidation is coupled to HCF(III) reduction. In the abiotic reactor, oxidation of HCF(II) is coupled to water splitting. HCF couple appears to be an interesting opportunity as a mediator in electrolyser for several reasons (Fig. 2): i) a low overpotential at low current density which will overcome MFC system limitation using oxygen reduction reaction; ii) a low thermodynamic redox potential which reduces energy costs for water splitting (vs anodic water oxidation); iii) a high reversibility and stability which justifies its wide use in electrochemical system.

The aim of this work is to validate the feasibility of such coupled systems as a proof of concept. Using a simple architecture, our MFC cascade system fed with glucose produces a current density up to 1.9 A m⁻² (with a COD removal rate of 30 mol m⁻² d⁻¹ and a HCF(III) reduction rate of 8.9 mol m⁻² d⁻¹). Thanks to this architecture (Fig. 1), we can lead an hydrogen production electrolysis at cell potential below 1 V with a current density up to 25 mA cm⁻² (250 A m⁻²) and a HCF(II) oxidation rate of 60 mol m⁻² d⁻¹. Regeneration rate of HCF in the electrolyser is about an order of magnitude higher, than in MFC cascade fed system.

Materials and methods

Electrolyser

In the electrolyser, two reactions occur at platinum electrodes, the HCF(II) oxidation in alkaline solution (Equation (3)) and the reduction of proton in sulphuric acid solution (Equation (4)).

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