



A wireless sensor powered by a flexible stack of membraneless enzymatic biofuel cells



Denis Desmaële^{a,*}, Louis Renaud^b, Sophie Tingry^a

^a Institut Européen des Membranes, UMR 5635, ENSCM-UMII-CNRS, place Eugène Bataillon, 34095 Montpellier, France

^b Université de Lyon, Institut des Nanotechnologies de Lyon INL-UMR5270, CNRS, Université Lyon 1, Villeurbanne F-69622, France

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ABSTRACT

This paper presents a novel multilevel membraneless enzymatic biofuel cell. For the first time, we introduce the use of thin polyester films as flexible electrode substrates for a Glucose/O₂ microfluidic biofuel cell. More specifically, we report a proof-of-concept based on a three dimensional (3D) microfluidic chip fabricated via rapid prototyping where two T-shaped microchannels are vertically stacked one above the other. Via the integration of two in-reservoirs, only three access ports are required and an even repartition of laminar flows is ensured within each microchannel. Both channels have gold electrodes patterned on their top and bottom walls. With four anodes/four cathodes having an individual area of 25 mm², our structure is only 425 μm thick. Enzymatic reactions occur via the use of glucose oxidase and hexacyanoferrate flowing in the anolyte solution whereas freely suspended laccase and 2,2-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid) diammonium salt (ABTS) are introduced in the catholyte stream. Under a flow rate of 150 μL min⁻¹ and with all anodes/cathodes connected in parallel, our structure can generate a maximum net power of 12.5 ± 0.05 μW. Connected to a voltage boost converter increasing the nominal output voltage to 3.1 V, we demonstrate that our proof-of-concept can already be exploited to supply electrical energy to a wireless sensor sending temperature measurements to a remote computer.

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1. Introduction

Enzyme catalyst biofuel cells are today recognized as promising alternative energy sources to act as miniature power supplies for low power electronic devices [1,2]. Conventionally, glucose biofuel cells architectures are similar to those of classical fuel cells. In particular, they often require a physical barrier (e.g., a proton exchange membrane) to separate the anodic and cathodic compartments [3,4]. In 2002, however, Ferrigno et al. reported the concept of a membraneless fuel cell where the properties of the laminar flow regime were exploited to demonstrate that fuel and oxidant streams can flow side by side without significant mixing in a microfluidic channel [5]. This concept eliminates the need for a physical barrier because the anode and cathode reactions are separated by the laminar flow, but protons are still allowed to transport across the fluid–fluid interface. Such a configuration hence simplifies the fabrication process and reduces costs. Furthermore, membraneless fuel cells are inherently well adapted to miniaturization since they can be produced via processes derived

from the microelectronic industry that provide high capabilities of integration.

Since the pioneering work of Ferrigno et al., different types of membraneless fuel cells using various reactants have been reported (see [6–9] for extensive reviews). The number of works dealing with membraneless designs of enzymatic glucose biofuel cells (EG-BFCs), however, still remains limited. Configurations involving enzymes immobilized on the surfaces of electrodes were reported in [10–12]. In [13], a multi-level structure featuring advanced functionalities was presented in order to prolong the performance of immobilized enzymes over a longer period of time. As an alternative approach, results involving bare electrodes and enzymes flowing in solutions were reported in [14,15]. Regardless of whether the enzymes were immobilized or not, all these membraneless EG-BFCs were produced by soft lithography, namely microchannels were replicated in Polydimethylsiloxane (PDMS) from master molds. Microchannels were then aligned and bonded to glass substrates with electrodes patterned via photolithography. Although PDMS is convenient to produce prototypes for the research laboratory, all these membraneless EG-BFCs were thick and rigid. Recently, a fabrication process based on a lamination method allowed to create a membraneless EG-BFC whose thickness was only 1 mm [16]. Notwithstanding the elegance of the approach, it is still needed to

* Corresponding author. Tel.: +33 04 67 14 91 57; fax: +33 04 67 14 91 19.

improve the net power that can be produced by membraneless EG-BFCs. Indeed, most polarisation curves found in the literature are normalized by the electrode surface so that the efficiency of different designs can be more easily compared. The performance of a membraneless EG-BFC is hence often presented in terms of current density (expressed in $A\text{ cm}^{-2}$) and/or power density (expressed in $W\text{ cm}^{-2}$). In conventional membraneless fuel cell designs, current and power densities are improved with small electrodes [17,18]. Similarly, membraneless EG-BFCs often tend to integrate electrodes with an area ranging from 4 mm^2 to 20 mm^2 . On an engineering point of view, however, the net power (expressed in W) proves to be the primary metric of interest [19]. With small electrode surfaces, the maximum power that can be generated is necessarily restricted. Nonetheless, extending the electrode surfaces without sacrificing the initial performance of a given design is not trivial. Depletion zones indeed tend to develop [14,17,18]. Even the integration of several small electrodes within the same microchannel can lead to altered performance [14]. The most straightforward way to increase the voltage and/or current generated by a membraneless EG-BFC is to replicate its design and connect electrodes that are contained in separate microfluidic cells. Some array configurations connecting several electrodes in series and/or in parallel were introduced in [20,21]. Such arrays can be seen as 2D configurations since the electrodes and fluidic cells were duplicated side by side on the same glass substrate. Nonetheless, such 2D approaches would yield devices with large footprint when considering the duplication of numerous electrodes/cells.

Therefore, we believe that it remains presently challenging to increase the maximum power of a membraneless EG-BFC by duplicating its design in an efficient and compact manner. In this paper, we present the preliminary results of our current research efforts to alleviate this limitation. In particular, we introduce a novel three dimensional (3D) and flexible architecture based on the vertical stacking of two microfluidic membraneless EG-BFCs. For the sake of conciseness, we call this new structure “flexible stack of membraneless fuel cells”, which will be referred to as FS-MFCs. Our FS-MFCs exhibits a high degree of flexibility and can be entirely assembled via a low cost, scalable fabrication process which is

entirely based on rapid prototyping. To illustrate its potential for concrete low power electronics, we demonstrate its capability to supply electrical energy to a wireless sensor transmitting temperature values to a remote computer.

2. Materials and methods

An overview of the complete experimental setup is given in Fig. 1. Details about each element are provided in the following paragraphs.

2.1. Chemicals and solutions

As detailed in Section 2.2, the FS-MFCs is a multi-anode/cathode structure. In order to facilitate its assembly, we decided to use enzyme catalysts flowing in solutions for our first tests. All chemicals were obtained from Sigma-Aldrich and used as received without further purification. Anolyte and catholyte were prepared based on protocols previously reported by our group. Additional details can be found in [22,23]. Briefly, glucose is oxidized to generate gluconolactone at the anodes whereas oxygen is reduced to water at the cathodes. The anolyte consisted of stock solutions of glucose (10 mM) prepared in neutral phosphate buffers and allowed to mutarotate overnight before use. Glucose oxidase from *Aspergillus niger* (1 mg mL^{-1} , $198,000\text{ U mg}^{-1}$ solid) and hexacyanoferrate (10 mM) were then dissolved in 10 mL of glucose solution. For the catholyte flow, 2,2-azinobis (3-ethylbenzothiazoline-6-sulfonate) (1 mg mL^{-1}) and laccase from *Trametes versicolor* (1 mg mL^{-1} , 20 U mg^{-1} solid) were prepared in 10 mL of 0.1 M citrate buffers (pH 5.0). Before starting experiments, the anolyte was purged with azote and the catholyte was saturated with oxygen for at least 15 min. Solutions were then transferred in 10 mL capacity syringes (BD Discardit II) and injected into the FS-MFCs using a syringe pump (KDS200, KD Scientific).

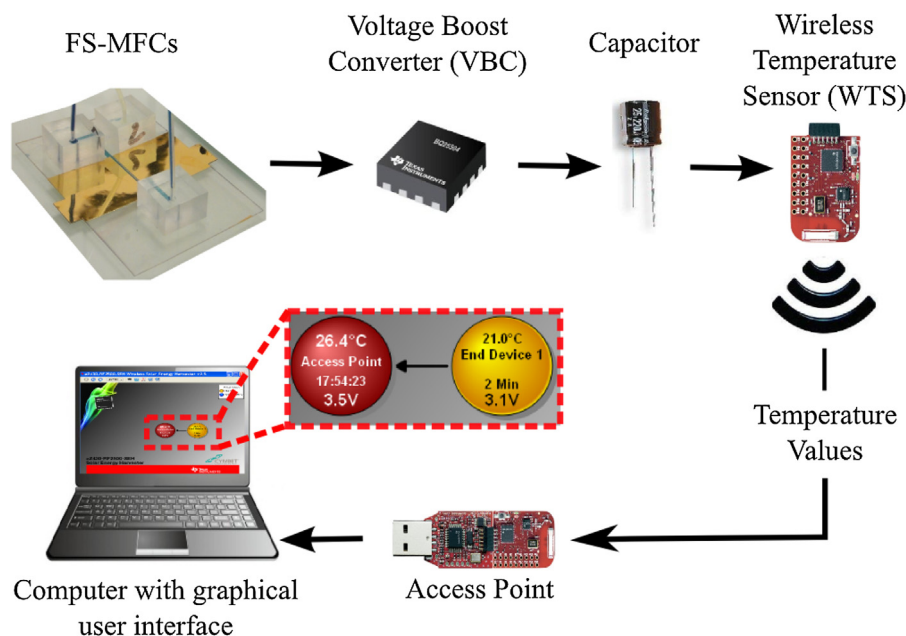


Fig. 1. Overview of the complete experimental setup tested in this paper. Because the FS-MFCs presented cannot be directly connected to a WTS, a VBC is first used to increase the nominal voltage to 3.1 V. The electrical energy generated over time by the FS-MFCs is then continuously stored in a capacitor. This configuration allows to sustain the bursts of power required by the WTS for periodically transmitting a temperature measure to an access point connected to a remote computer.

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