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Research paper

A laminar-flow based microbial fuel cell array

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ABSTRACT

We created a novel laminar-flow based microbial fuel cell (MFC) array to be an integrable and scalable power source for portable lab-on-a-chip (LOC) devices. The microfluidic MFC enabled the laminar flow of anolyte and catholyte streams in a microchannel without any physical membranes while the device harvested electricity by utilizing ion transfers through the laminar interface which acts as a virtual membrane. The array prototype incorporated four series-connected fuel cells and was operated with two common inlets for the continuous introduction of the anolyte and catholyte. In the anodic flow region, microorganisms oxidized organic media and completed respiration by transferring the electrons to the anodes. The protons generated by the anodic reactions passed through the liquid-liquid interfaces and traveled to the cathodic streams. The electrons then moved across the external resistors to the cathodes where they combined with the protons and reduced oxidant (i.e. catholyte). The array generated a maximum power output of 60.5 $\mu W/cm^2$ using a 100 kΩ load, which outperformed a single laminar flow MFC unit by a factor of approximately 4. The series or parallel application of this array structure, using microfluidic MFCs integrated into a single LOC device, can offer the potential for on-chip power generation.

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1. Introduction

Portable lab-on-a-chip (LOC) devices have recently gained increasing attention as a new platform for point-of-care (POC) diagnostics [1–3]. One of the key challenges in developing POC devices is to effectively and sufficiently power them [4]. Especially in resource-limited environments, where the electricity grid is not developed, and batteries are not available, power supplies are one of the most critical components for stand-alone and self-sustained POC devices [5–7]. Accordingly, the demand for integrating microscale power sources into LOC devices has been rapidly expanding.

Laminar flow-based fuel cells (LFFCs) have recently emerged as a new type of the microscale power supply for those LOC devices [8–11]. The LFFCs operate by using microscale, laminar, and parallel streams of fuel (i.e. anolyte) and oxidant electrolytes (i.e. catholyte) without a physical membrane to separate the anodic and cathodic compartments. This simple device configuration overcomes many limitations of fabrication, performance, and operation of the fuel cells [12–15].

Recently, this LFFC technique was applied to the design of biofuel cells such as microbial fuel cells (MFCs) [16–19], where the bacterial media and oxidant streams are naturally separated, so that the proton exchange membranes are no longer needed (Fig. 1a). The use of the physical membranes has been a major limitation in developing a cost-effective and simplified MFC device because the membrane inclusion results in a substantial increase of material/fabrication cost and significant voltage loss during the charge transfers through the physical membrane [17,20]. The laminar flow platform can be a potential solution to current challenges associated with the development of the microscale MFCs, providing unique advantages over conventional designs such as simplified device architectures, lower costs, and easy integration with lab-on-a-chip technologies [16–20]. Moreover, the membrane-less configuration increases the MFC performance with a reduction of internal resistance and an improvement in mass transport [17]. Lastly, the microscale characteristic lengths of microfluidic chambers enable short start-up times for bacterial accumulation and acclimation on the anode and ultimately rapid power generation of the MFCs [19].

Despite this vast potential, however, the application of the laminar flow-based MFCs (LFMFCs) as a power supply is still severely limited because of their low performances associated with the inherent device size. The typical sustainable voltage output from

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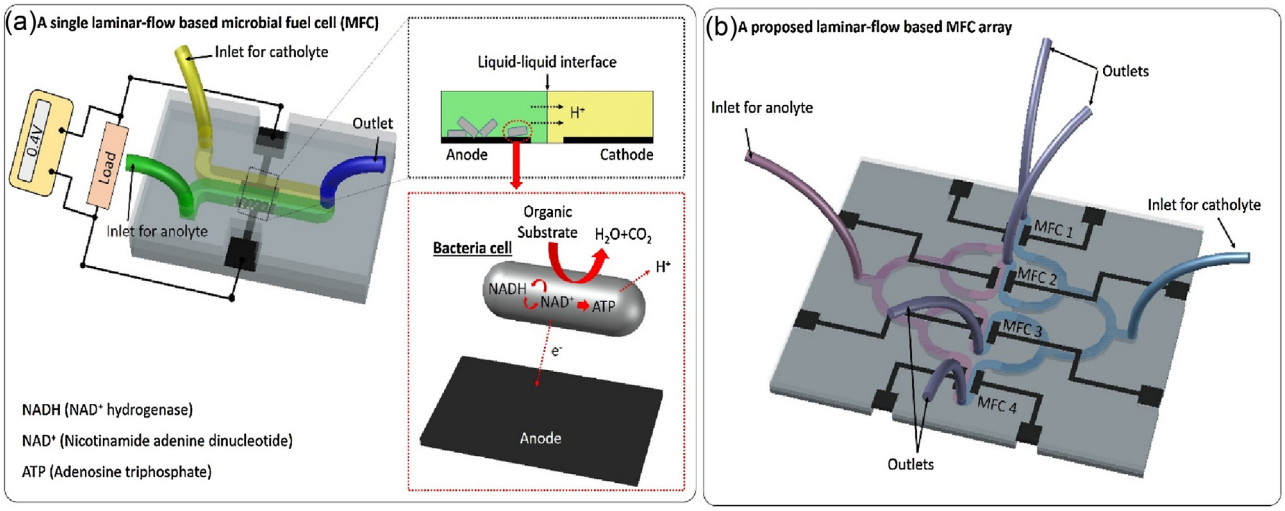

Fig. 1. (a) A single laminar-flow based microbial fuel cell (MFC). The electrodes, bacterial inoculum (i.e. anolyte), and oxidant (i.e. catholyte) are all contained in a microfluidic channel and are operated without the use of a physical barrier to separate the anolyte and catholyte streams. Bacteria oxidize organic matter in the anolyte and transfer the electrons for their anodic respiration. During the process, NAD+ and NADH function repeatedly oxidize and reduce to synthesize ATP. (b) A laminar-flow based MFC array, as proposed in this work. The array incorporates four MFCs and operates with two common inlets for the continuous introduction of the anolyte and the catholyte, respectively. The four units can be connected electrically in series to produce higher operating voltage and power output.

a single microfluidic MFC unit is on the range of 0.3–0.8 V, which limits their applications only to a laboratory tool, used to screen electroactive microorganisms and guide the development of large-scale MFC systems [17,19]. To produce sufficient voltage (1.5 V) and/or power (to reside within the operating range of electronics), it is therefore necessary to either scale up one single unit or connect multiple small units in series. From our preliminary results [21,22] and literature survey [23–25], it is well known that a large number of units connected together will produce more power than a single unit and more suitable for a LOC device with a small microfluidic device footprint. To date, however, there have been no attempts to connect multiple MFC units together in microfluidics. The working hypothesis of this work was that microfluidic MFC scale-up could be achieved by connecting multiple miniature units in a single microfluidic chip and the fluidic operation could be easily manipulated by a network of branched microfluidic channels with only two common inlets for the continuous introduction of anolyte and catholyte, respectively (Fig. 1b).

2. Experimental procedure

2.1. Operating principle of LFMFC array

Generally, MFCs consist of anodic and cathodic chambers separated by a proton exchange membrane so that only protons or other cations can move from the anode to the cathode. An external resistor connects the two electrodes to complete the external circuit. Microorganisms oxidize organic matter in the anodic chamber, completing respiration by transferring electrons to the anode [20]. During this process, chemical energy is captured throughout the electron transport chain. Nicotinamide adenine dinucleotide (NAD+) and nicotinamide adenine dinucleotide dehydrogenase (NADH) function as coenzymes for the reactions, repeatedly oxidizing and reducing to synthesize adenosine triphosphate (ATP), the biological energy unit (Fig. 1a) [26,27].

In the laminar flow-based MFCs (LFMFCs), the microbial metabolism at the anode and the reduction process at the cathode lead to the conversion of chemical energy into electrical energy while the protons generated from the anodic reaction can cross the virtual membrane (i.e. the liquid-liquid barrier) to maintain the electro-neutrality of the MFC system [17,19]. At the anode, electrons are extracellularly transferred to the anode and flow to the cathode through the external electrical circuit. At the cathode, ferricyanide, $[Fe(CN)_6]^{3-}$, captures the electrons and the reduction reaction is completed as follows.

$$[Fe(CN)_6]^{3-} + e^- \rightarrow [Fe(CN)_6]^{4-} \quad (1)$$

Four LFMFCs were integrated into an array with parallel fluidic connections through Y-shaped micro-channels (Fig. 1b). Two common inlets were divided into four full operating fuel cells creating well-separated electrode pairs with the virtual laminar boundaries. The four units were connected electrically in series to produce a higher operating voltage and power output.

2.2. Device fabrication and assembly

As shown in Fig. 2a and b, the LFMFC array features Y-shaped microfluidic chambers with four pairs of electrodes. The size of the array is 5.5 cm × 7 cm with four MFC units (8 mm long, 3.8 mm wide, and 102 μm thick (Fig. 2b)). The LFMFC array was developed by sandwiching four layers (Fig. 2c); (i) a bottom poly(methyl methacrylate) (PMMA) layer, (ii) a PMMA layer with multiplexing fluidic channels, (iii) a 102 μm-thick thin plastic film (Vinyl-Pane) with Y-shaped channels, and (iv) a top PMMA layer with carbon electrodes (1 mm × 5 mm) with holes for common inlets and four outlets. Each layer was micro-patterned by using laser micromachining (Universal Laser Systems VLS 3.5). We used one of the most common electrode materials for microorganism-based fuel cells, a carbon-based material, which provides a large surface area and functional organic groups, favoring cell vitality [20]. The carbon (Graphite ink, Ercon Inc.) was deposited on the pre-defined area of the top PMMA layer through screen/stencil printing and dried on the hot plate at 95 °C for 15 min. The four layers were manually stacked in sequence to form the channels and expose the electrodes to the future fluidic paths, while carefully aligning the tubing holes. All the layers were thermally bonded at 140 °C for 1 h with pressure applied by binder clips. Fluidic tubes (CODAN, 0.35 mL volume) for two inlets to the array and four outlets from individual MFC units were plugged into the holes completing the laminar flow paths (Fig. 2a). The assembled device was sterilized with 70% ethanol and then ultraviolet light for 24 h.

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