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Toward the scaling up of microfluidic fuel cells, investigation and optimization of the aggravated cathode flooding problem

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ABSTRACT

The scaling up of microfluidic fuel cells (MFC) with enlarged electrode areas can be very advantageous for their power output and further stacking, which may, however, aggravate the cathode flooding problem due to intensified fuel crossover. In this paper, we first validate this phenomenon in a scaled-up MFC prototype, which has a hydrogen-breathing anode and an air-breathing cathode. Possible influential factors on the flooding extent are studied next, including the electrode area, electrolyte flow rate, and ambient temperature. Furthermore, three different strategies are proposed to improve cathode flooding, which is achieved either by adding an interlayer inside the channel to suppress hydrogen crossover, by adding hydrophilic water remover on the cathode surface to wick the generated water, or by cracking the cathode catalyst layer (CL) to manipulate the water transport direction. Among all these methods, the cracking of cathode CL is found to be remarkably effective especially with acid electrolyte, which exhibits the lowest current density degradation rate of 1.3 mA cm^{-2} h^{-1} when discharged at the peak power point for five hours.

1. Introduction

Microfluidic fuel cells (MFC), also known as membraneless fuel cells or laminar flow fuel cells, have attracted extensive research and development efforts in recent years [1–4]. As its name suggests, a MFC employs microfluidic flows as electrolyte instead of any solid membrane electrolyte, with all its cell components confined in a micro-channel. Benefited from the elimination of membrane, many distinct advantages against conventional membrane-based fuel cells are obtained in MFCs, including cost efficiency, fuel and oxidant flexibility, adjustable pH of the reaction environment [5–7], inherent heat and water management, etc. As shown in Fig. 1, there are three main types of flow configurations for MFCs in the literature, namely the co-flow, the counter-flow, and the single-flow configuration [8]. A co-flow MFC generally utilizes two laminar flows, i.e. the anolyte and the catholyte, in parallel to deliver the dissolved fuel and oxidant to the anode and cathode, respectively [9]; while a counter-flow MFC delivers the two flows face to face passing the electrodes, before they come across and flow together to the exit [10–12]. In addition, the single-flow MFC with simpler fluid management has also been proposed, which is especially suitable for gaseous fuels such as hydrogen [13,14] or methanol vapor [15]. Till now, satisfactory cell performance has been achieved in MFCs with various combinations of fuel and oxidant, which is comparable with or even superior than their membrane-based counterparts [7]. Nevertheless, their absolute power output is very limited due to their tiny electrodes. Therefore, it is necessary to stack multiple MFC single cells together, in order to achieve sizeable voltage and power output for practical applications [16,17].

Restricted by the micro-channel, electrode size in present MFCs is generally in the order of several mm^2. As a consequence, a large number of MFC single cells need to be stacked together to fulfill practical power demands, which inevitably leads to sophisticated fluid management and great system complexity. In addition, the stacking efficiency will also be dragged down due to the increased shunt currents between adjacent single cells [16,17]. Furthermore, operational stability of such systems is questionable, as the failure of one single cell will cause a chain-effect to the others due to their unavoidable fluid connection. In consideration of all these, the scaling-up of MFC single cells with enlarged electrode area is highly promising, which requires a much reduced amount of single cells in the stack for a specific power demand. However, for the co-flow and counter-flow MFCs, enlargement of electrodes will bring intractable problems, including intensified depletion of boundary layer on the electrode surface, less stability of the flow interface

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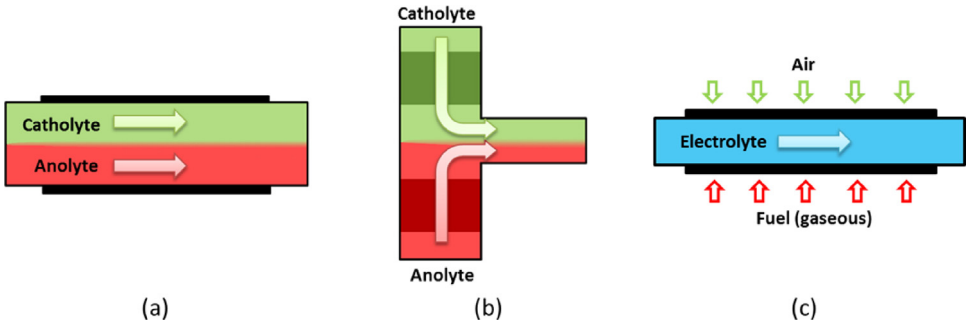

Fig. 1. Three different flow configurations of MFC: (a) co-flow; (b) counter-flow; (c) single-flow.

between the two streams, and severer crossover problem. As for the single-flow MFC, depletion of boundary layer and flow interface no longer exist benefited from its distinctive flow configuration [15], which makes it especially suitable for scaling up purpose. Nevertheless, fuel crossover will still be intensified due to the inevitable enlargement of channel size, which may cause the aggravated cathode flooding problem.

Cathode flooding is a common issue in low-temperature fuel cells, such as the proton exchange membrane fuel cell (PEMFC) [18,19]. The accumulated water in the gas diffusion electrode (GDE) will have negative effects on cell performance due to the elevated mass transport resistance, which is especially associated with the cathode side where water is continuously generated via both the oxygen reduction reaction (Eq. (1)) and the oxidation of cross-overed H_2 (Eq. (2)), as shown in the following chemical equations:

$$O_2 + 4H^+ + 4e^- \xrightarrow{Pt} 2H_2O \quad (1)$$

$$2H_2(crossovered) + O_2 \xrightarrow{Pt} 2H_2O \quad (2)$$

If not removed in time, the generated water will accumulate inside the porous cathode first, and finally emerge on the cathode gas diffusion layer (GDL) surface, i.e. the flooding occurs. To tackle with this issue, many research works have been done with PEMFCs, including flow channel design [20–22], hydrophilic wicking on cathode surface [23], modification to catalyst layer (CL) [24] or GDL [25], electro-osmotic pumping [26–28], etc. As for MFCs, cathode flooding can be greatly alleviated by the continuous electrolyte flow, which can not only suppress fuel crossover, but also remove the generated water from the CL efficiently [29]. As a consequence, water is less likely to accumulate, leading to unobstructed gas passages for oxygen supplement. However, as for scaled-up MFCs, since fuel crossover is intensified, whether cathode flooding can still be well controlled remains to be seen, which has not been studied yet to the best of our knowledge.

In this study, a scaled-up MFC prototype with H_2-breathing anode and air-breathing cathode was utilized to study the cathode flooding issue. The phenomenon of cathode flooding was first verified experimentally, proving that flooding can also occur in scaled-up MFCs. Next, a parametric study was conducted to investigate possible influential factors on the flooding extent, including electrode area, electrolyte flow rate, and ambient temperature. Finally, three different optimization strategies for flooding control purpose in scaled-up MFCs were proposed and testified.

2. Experimental

2.1. Fuel cell fabrication

As shown in Fig. 2, the present scaled-up MFC prototype was mainly composed of two parts, i.e. the fuel cell part and the H_2 flow field part, which were combined together by six pairs of bolts&nuts in their periphery and a silicon rubber gasket in between. The fuel cell part had a multi-layer structure, including four poly methyl methacrylate (PMMA) layers, three silicon rubber layers, two GDEs, and two silver foil curent collectors. In its middle, a 0.5 mm-thick silicon rubber layer with a mico-channel (Channel layer) was sanwiched and sealed by two 0.5 mm-thick PMMA layers which

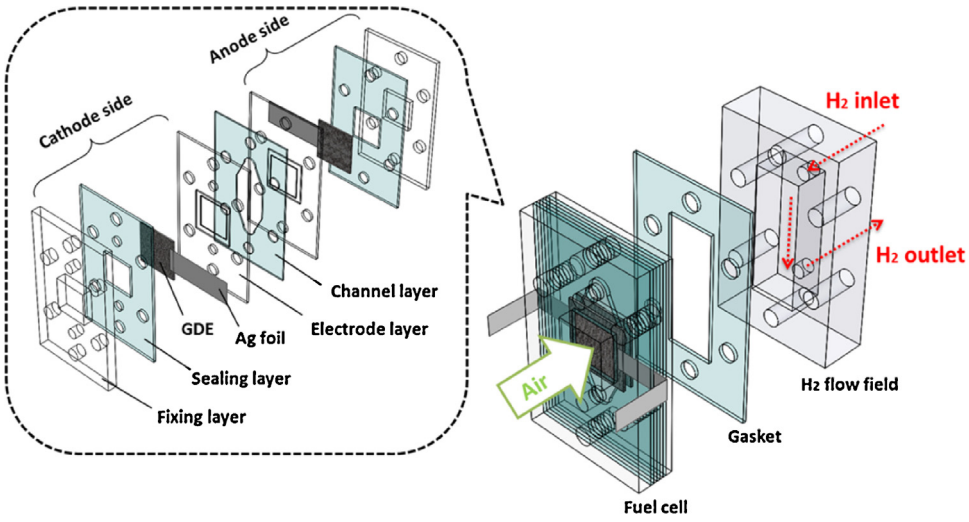

Fig. 2. Composition of the present scaled-up single-flow MFC prototype.