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Effect of serpentine flow-field design on the water management of polymer electrolyte fuel cells: An in-operando neutron radiography study

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HIGHLIGHTS

- In-depth understanding of water dynamics through in-operando neutron radiography.
- Single-, double- and quad-channel serpentine cathode flow-fields compared.
- The single-channel exhibits better performance and water uniformity.
- Three regimes identified: increasing hydration, flooding and drying out.

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ABSTRACT

In-depth understanding of the dynamics of water formation, accumulation and removal is important for flow-field design optimization to ensure robust performance and durability of polymer electrolyte fuel cells (PEFCs). Here, in-operando neutron radiography is used to display and quantify liquid water distribution across the entire active area of single-, double- and quad-channel serpentine flow-fields. The results revealed that the water management and performance of PEFCs is strongly affected by the number of serpentine channels in the cathode flow-field. The single-channel serpentine-based PEFC exhibits both a better cell performance and uniformity in the local water distribution. The quad-channel based PEFC exhibits the largest voltage fluctuations caused by severe water flooding in the gas channels. However, the single-channel design leads to significantly larger pressure drop than the multiple-channel counterparts, which requires much higher parasitic power to pressurize and recirculate the reactants.

Three different regimes of operation can be defined based on the current density: gradually increasing hydration ($< 400\ mA\ cm^{-2}$), flooding ($400\ mA\ cm^{-2} \leq j \leq 600\ mA\ cm^{-2}$) and drying out ($>600\ mA\ cm^{-2}$). The reduced overall quantity of water in the channels with an increase in current density can be attributed to faster gas velocity and higher cell temperature.

1. Introduction

Polymer electrolyte fuel cells (PEFCs) carry advantages of high electrical efficiency, low operating temperature and rapid start-up, which are desirable for a wide range of applications [1,2]. A key element that dictates PEFC performance and durability is water management [3–5]. On the one hand, the membrane (typically Nafion) needs to be well-hydrated to achieve high ionic conductivity and prevent the membrane from drying out, which can lead to increased Ohmic and activation losses [6]. On the other hand, excessive water can impede effective reactant transport and lead to flooding, which results in performance decay and cell failure [7–9]. Thus, a careful water balance is required for robust PEFC performance.

Flow-fields are crucial components that affect the water management and performance of PEFCs. They distribute the reactants over the active area and remove excess water generated from electrochemical

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reactions. An improper flow-field design will cause maldistribution of water, which affects performance and lifespan [10–14].

A range of flow-field designs have been proposed, including parallel [15], serpentine [16–18], interdigitated [19], bio-inspired [13,20,21], pin-type [22], and cascade flow-fields [23]. Serpentine is the most commonly employed designs in the cathode for its superior water management capacity. The long channel length of the serpentine configuration leads to greater pressure drop, which can alleviate water build-up in channels and enhance convection under the rib/land. Consequently, PEFCs using serpentine flow-fields at the cathode tend to have better water management and performance [18,24,25], but the greater back-pressure requires more effort to deliver the reactant, resulting in a larger parasitic load burden.

Serpentine flow-fields are classified according to their channel numbers; that is the number of parallel channels that trace a common serpentine pathway. Modelling studies have examined the effect of channel number, and showed that the current density, temperature and produced water tend to be more uniform with the increment in the channel numbers [26]. Computational fluid dynamic (CFD) simulations indicate that double-channel serpentine yields more uniform liquid water distribution than the single-channel serpentine [27]. However, an opposite trend was reported by Limjeerajarus et al. [28] and Wang et al. [29], which indicates that, for a small-sized PEFC, the single-channel serpentine flow-field provides both better water uniformity and related cell performance, attributed to the larger pressure drop caused by its long path length, leading to enhanced water removal.

The reliability of these simulation results requires appropriate experimental validation that includes characterisation of internal water management. To this end, a range of diagnostic techniques have been employed. Technique such as optical visualisation is a powerful means of determining liquid water production and removal within the flow channels [30,31]. However, the PEFCs need to be modified since a transparent window is required for observation, which increases the complexity of the cell design and renders the cell unrepresentative of a practical system. Localised electrochemical measurements can be used to infer the state of water within a PEFC. Measurement of the Ohmic resistance indicates the level of hydration of the membrane, which can be measured using high-frequency impedance (HFR) [32]. The pressure drop across the inlet and outlet of the reactant is an indicator of the water management capacity of flow-field designs [13,26–29]. Larger pressure drop results in effective convective liquid water removal from the cell. On the contrary, it requires higher parasitic power to pressurize and recirculate of the reactants.

Of the reported PEFC water mapping methods in the literature, neutron imaging is arguably the most powerful. The high attenuation of neutrons by liquid water [33] and high transparency of neutrons to other PEFC components allow for high-resolution measurements of liquid water formation and transport. First pioneered by Mosdale et al. in 1996 [34], researchers have mainly focused on applying this technique to visualise water in a PEFC under different current density, temperature, stoichiometry, and reactant humidification conditions [9,35–39].

However, relatively few works employ neutron imaging to study the significant effect of flow-field designs on the water management of fuel cells. Limited existing works related to this issue examined the effect of channel geometric parameters (such as depth, corner angle and land/channel ratio) on the water management of PEFCs [40–42]. On the contrary, there is no other report in the literature providing an in-depth study of the effect of water distribution in a fuel cell as a function of the serpentine channels number. As the most commonly employed designs in the cathode of PEFCs, the investigation of the effect of serpentine flow-field design on the water management is indispensable to deliver performance and lifespan improvements in PEFCs. Therefore, our study fills a gap in the literature and for the first time, provides a systematic comparison of the water management in different serpentine flow-field designs.

In the present study, by combining neutron imaging with pressure drop measurement and monitoring the high-frequency impedance, it is possible to explore the influence of different serpentine flow-field designs on water management and related PEFC performance. The comparison of different flow-field designs was made by keeping geometric parameters of the channels constant in order to isolate the effect of cathode channel number on fuel cell performance.

2. Experimental setup

2.1. PEFC design

A closed-cathode PEFC with an active area of 9.5 cm^2 was designed. The cell consisted of two printed circuit board (PCB) flow-fields, a membrane electrode assembly (MEA), gaskets, and two end-plates. The anode and cathode PCB flow-fields were 0.8 and 1.6 mm thick, respectively. PCB-based fuel cells have the advantage of being cheap, light-weight, and easily customisable to specific design requirements; they have been employed extensively for current and temperature distribution measurements [31,43,44] and are being commercialised by the likes of Bramble Energy in the UK.

The copper layers of the PCB were electroplated in nickel and gold to prevent corrosion. The anodic flow-field features a single serpentine flow channel with channel width and spacing of 1 mm, and depth of 0.5 mm (Fig. 1(a)). Three cathode serpentine flow-fields with single, double, and quad channels were fabricated to study the influence of serpentine channel numbers on water management at the cathode (Fig. 1(a)). The width of land and channel, and the channel depth are 1 mm.

A 70 μm thick sheet of Tygaflor was used as gasket at the interface between flow-fields and end-plates. The same material was used as gasket to seal the perimeter of the MEA. The MEA was fabricated in-house by hot pressing the Nafion 212 membrane (DuPont, USA) and ELE00162 gas diffusion electrodes (Johnson Matthey, UK). The MEA was pressed at 130 °C for 3 min with an applied pressure of 400 psi. The anode and cathode aluminium end-plates were 8 and 10 mm thick, respectively, and were electroless plated in gold to prevent corrosion. An exploded view of the fuel cell assembly is shown in Fig. 1(b).

2.2. Experimental procedure

An in-house designed LabVIEW software (National Instruments, USA) was used to control the PEFC operation (air, hydrogen, and the load) and record the data through communicating with a data acquisition card (DAQ card, USB 6363, National Instruments, USA). The PEFC was operated at ambient temperature in absence of reactant humidification. The stoichiometry of cathode and anode flow was kept at 3 and 1.2 across the investigated current density range, respectively. This leads to faster gas velocity at higher current density, which fosters effective convective liquid water removal in the channel. Gas was supplied in counter-flow orientation in order to enhance membrane hydration [45], where hydrogen is fed from the bottom right of the cell and air is fed from the top left of the cell (Fig. 1(a)). The flow rates of inlet gases were controlled using digital mass flow controllers (Bronkhorst, UK). The pressure drop across the cathode was monitored using pressure transducers (Variohm, UK). The high frequency resistance (HFR) of the PEFCs during operation was measured via electrochemical impedance spectroscopy (EIS) to gauge the hydration level of the membrane. Current was drawn from the fuel cell using an electronic load (PLZ664WA, Kikusui). Current sweep experiments were carried out in steps of 200 mA cm^{-2} for 10 min per point from 0 to