Electro-thermal mapping of polymer electrolyte membrane fuel cells

with a fractal flow-field

V. S. Bethapudi a, b, J. Hack b, G. Hinds c, P. R. Shearing b, D. J. L. Brett b, *, M. –O. Coppens a,

**

a EPSRC “Frontier Engineering” Centre for Nature Inspired Engineering & Department of
Chemical Engineering, University College London, London WC1E 7JE, United Kingdom
b Electrochemical Innovation Lab, Department of Chemical Engineering, University College
London, London WC1E 7JE, United Kingdom
c National Physical Laboratory, Hampton Road, Teddington, Middlesex TW11 0LW, UK

Abstract

Electro-thermal maps of a polymer electrolyte membrane fuel cell (PEMFC) show the spatial
distribution of current density and temperature, which is useful to evaluate their
performance. Here, electro-thermal mapping is carried out for the first time on a PEMFC with
a fractal cathode flow-field, the design of which emulates the efficient, scalable air transport
inside the lungs. Such maps are compared with those of a conventional single-serpentine
flow-field PEMFC. Each cell’s performance is characterised by analysing the surface
distribution of current density and temperature at different reactant relative humidity (RH)
and cell voltage. Relationships are shown between segment current densities and surface
temperatures, and between reactant relative humidity and cell operating conditions. The cells
with a fractal flow-field deliver better electrochemical performance and exhibit more
homogeneous current distributions compared to those with a single-serpentine flow-field, in
which the current distribution is non-uniform due to cell flooding. The surface temperatures
are higher in cells with a fractal flow-field than in those with a single-serpentine flow-field, consistent with the observed cell performances. In addition, electrochemical impedance spectroscopy characterisation indicates flooding in the single-serpentine cells, but not in the fractal cells.

Keywords:

Current mapping; Temperature mapping; Fractal; Nature-inspired; Fuel cell; Flooding

* Corresponding author.

d.brett@ucl.ac.uk

Professor of Electrochemical Engineering

Department of Chemical Engineering

University College London

Torrington Place

London, WC1E 7JE

** Corresponding author.

m.coppens@ucl.ac.uk

Ramsay Memorial Professor

Department of Chemical Engineering

University College London

Torrington Place

London, WC1E 7JE
1. Introduction

Polymer electrolyte membrane fuel cells (PEMFCs) operating on hydrogen, with no emissions at the point of use, are gaining prominence as alternatives to energy devices powered from fossil energy. PEMFCs supply uninterrupted power, as long as sufficient fuel (hydrogen) and oxidant (air) are supplied to support the electrochemical reactions. This is an advantage of PEMFCs compared to batteries, which cannot support continuous operation, due to their periodic charging needs. The electrical efficiency of PEMFCs can go as high as 60% and the overall efficiency can be further improved if the PEMFC is operated in combined heat and power mode [1,2]. For the successful and efficient functioning of PEMFCs, several components associated with their operation must work optimally. Of these, flow-fields play a vital role in the transport and the distribution of reactants to the electrodes, where the electrochemical reactions occur.

PEMFC flow-fields are categorised based on the geometrical configuration of their reactant flow channel path. The serpentine flow-field geometry is a commonly used channel configuration that provides reactant flow to the electrode surface, uniform stack compression and effective water and thermal management [3]. However, the serpentine flow-field has a relatively long reactant flow path that often results in concentration gradients and pressure drop along its length [4]. In addition, stagnation of liquid water along the serpentine flow paths can occur, resulting in mass transfer-related issues, such as channel flooding and reactant starvation that reduce the overall cell performance [5,6].

Lung-inspired, fractal flow-fields ("fractal cells" for short) have been identified to deliver better performance (higher electrical power) compared to those with conventional single-serpentine flow-fields, while overcoming the aforementioned issues related to flow
The design of lung-inspired flow-fields [7–10] is based on rigorously proportioned, scale-invariant structural features that lead to optimally efficient air transport inside lungs, as opposed to biomimetic designs that are developed either by copying apparent features of biological transport structures, such as veins, lungs and leaves, or by integrating flow mechanisms observed in nature into conventional flow-field geometries, like serpentine, interdigitated and parallel configurations for air transport [11–13]. Theoretical studies on lung-inspired flow-fields by Kjelstrup, Coppens, Pharoah and Pfeifer [7] predicted that a 2D planar fractal flow distributor could more uniformly distribute air over the membrane-electrode assembly (MEA) region, compared to a serpentine flow-field, but also do so in a thermodynamically optimal way. This study did not simply mimic biology, but applied geometric features of the upper airway tree of mammalian lungs, which are scalable and commensurate with minimum entropy production, to design fractal flow-fields for fuel cell applications [7,14]. Trogadas et al. [8], Cho et al. [9], and Marquis et al. [15] carried out numerical simulations incorporating three-dimensional finite element models of the flow-fields and gas diffusion layers (GDLs), in combination with two-phase agglomerate models for the catalyst layers, to study the effects of the GDL thickness and the number of fractal branching generations on the PEMFC performance. These numerical simulations confirmed analytical predictions that the ideal number of branching generations, $N$, is between $N = 5$ and $N = 7$ for a flow-field plate with a surface area of 10 cm$^2$, such that convection-dominated flow and diffusion-controlled transport are balanced at both the outlets and the inlets (Péclet number, $Pe \approx 1$) [8,9]. Prototypes of lung-inspired cathode flow-fields with, first, 10 cm$^2$ and, then, 25 cm$^2$ effective MEA area were built on the basis of the computationally assisted designs discussed in [8,9]. These 3D branching fractal flow-field plates were fabricated using selective laser sintering of stainless steel [8]. It was shown that a fractal cell with $N = 4$
generations delivered a higher power density compared to a single-serpentine cell of the same MEA area at 50% and 75% reactant relative humidity (RH) conditions. It also demonstrated 50% lower values in pressure drop compared to the conventional single-serpentine flow-field design for all RH conditions tested. However, for both 10 cm² and 25 cm² cells, the fractal cell’s performance degraded considerably under 100% RH conditions, where additional water saturation occurred in the electrodes and the flow-field channels, resulting in mass transport issues due to flooding. In addition, when fractal cells having $N = 5$ generations were tested, water flooding was observed during operation, irrespective of the reactant conditions. This is due to ineffective water removal from the fractal cells, which limits their usage at higher reactant humidity. Furthermore, current disadvantages related to using selective laser sintering for flow-field manufacturing are the complexity and cost involved.

A novel method of 3D fractal flow-field development using 2D planar printed circuit board (PCB) plates with a layer-wise assembly approach by Bethapudi et al. [10,16] overcame the flooding issues and fabrication disadvantages associated with the previous designs [8]. Here, a fractal flow-field was developed, with air outlet paths modified from the previous design, which delivered superior performance to conventional single-serpentine cells under different operating conditions, even at 100% reactant RH. The performance enhancement in the fractal cell has been characterised and established by polarisation, temperature, galvanostatic and electrochemical impedance measurements.

Current and temperature mapping have been instrumental for the *in situ* diagnosis and analysis of various factors that affect the performance of fuel cells, such as water management [17–19], reactant concentration and distribution [5,18,20,21], operating conditions [22,23], flow channel configurations [24], thermal management [25–27] and cell
Some of the current distribution measurement techniques include indirect correlations based on local values, use of magnetic effects and Hall sensors, dependent on local potential measurements at the GDL and catalyst layer, and segmented measurement. Temperature distribution measurement techniques include thermocouple insertions, infrared imaging and segmented measurement. Of these, segmented measurements have been identified as a particularly powerful technique, especially for combined current and temperature measurements, due to their ability to characterise localised phenomena, such as reactant starvation and flooding inside the fuel cell. Furthermore, PCB-based segmented current collectors are widely used in the combined measurement of current and temperature, due to their low cost, flexibility during assembly and disassembly and ability to perform in situ measurements within cells.

As discussed, previous studies have identified the benefits of fractal fuel cell cells, compared with conventional single-serpentine cells. These studies were primarily dependent on using in situ electrochemical diagnostic techniques that established better hydration distribution and regulation inside the fractal cells, and their associated cell performance. In this study, design, development, and testing of a scaled-up layer-wise printed circuit board (PCB) based cathode fractal flow-field, compared to the previous PCB based lung-inspired flow-field with $N = 5$ fractal generations is presented. Simultaneous current and temperature mapping are used to understand the reasons for the improved performance and investigate the role of reactant RH and operating conditions, in addition to conventional electrochemical performance tests. Segment currents are evaluated and analysed to understand the impact of hydration distribution on the local currents generated in the cell, which provides new
insights and should help to guide further design, scale-up, and operation improvements of fractal fuel cells.

2. Experimental setup and characterisation methods

2.1 Flow-field design

The cathode fractal flow-field was developed using a layer-wise printed circuit board (PCB) plate technique, an approach that has been identified as cost-effective, easy and scalable for manufacturing [10]. Several layers of 2D planar PCB plate, with each plate consisting of a particular generation of the fractal flow-field structure, as shown in Fig. 1, were assembled to produce a cathode flow-field with 3D hierarchical, fractal geometry. The cathode fractal flow-field used here employed a 5-generation hierarchical fractal structure, with the airflow through this flow-field occurring from a single inlet to 1024 outlets, covering an effective MEA area of 25 cm². Each of the 5th generation outlets had dimensions of 400 μm × 800 μm, with a spacing of 1.18 mm between adjacent outlets. Furthermore, a surface vertical flow path of 0.5 mm in both width and depth ran through this spacing.
Figure 1: Schematic outline and order of 2D planar PCB plates (generations and interconnecting plates) used for the development of a 5-generation cathode fractal flow-field plate. Plate 1: gold-coated; plates 2 – 10: plain PCB plates.

These surface paths act as the flow outlet for reactant air and water generated in the cathode region, making this flow-field an open-ended cathode fractal flow-field. A total of 10 PCB plates were used, with 6 plates (1, 2, 4, 6, 8 and 10) accommodating the 5 generations of the fractal structures, and 4 plates (3, 5, 7 and 9) acting as interconnecting plates between generations, respectively (Fig. 1). Plate 1, which adjoins the MEA, was gold-coated and acted as the cathode current collector for this flow-field, while all other plates (2 — 10) were plain PCB plates. Similarly, the cathode single-serpentine flow-field was developed using PCB plates with a 1 mm² square cross-sectional area single-serpentine channel with a depth of 1 mm and acted as the cathode current collector. Additional plain PCB plates were introduced with the cathode single-serpentine flow-field to ensure that the overall thickness of the single-serpentine and fractal flow-fields remained the same. Furthermore, vertical flow paths having 0.5 mm width and 1 mm depth were provided in the cathode single-serpentine flow-field,
with a spacing of 1.18 mm between them, for the removal of reactant air and product, including crossover water. The effective MEA area covered by the cathode single-serpentine flow-field was 25 cm². The corresponding cathode fractal and cathode single-serpentine flow-fields are shown in Fig. S1.

The corresponding anode flow-field (Fig. 2) was constructed from a graphite plate of 3 mm in thickness. Here, graphite was used because of its high electrical and thermal conductivity, which facilitates current and temperature distribution measurements in the cells. The anode flow-field had a single-serpentine square channel with an area of 1 mm² and a depth of 1 mm.

Besides, the anode current collector used was a gold-coated PCB plate.

**2.2 Flow-field fabrication**

The different flow-field features discussed above were fabricated using a Roland-40 CNC setup (Roland, USA). In the cathode fractal flow-field, plate 1 had a 35 µm thick Cu coating that was modified by first electroplating Ni from 0.13 M Ni(NO₃)₂ solution at 4.3 mA cm⁻² (corresponding to between 3 V — 3.5 V applied voltage) for 3 min. It was then electroplated with Au from 0.02 M KAu(CN)₂ solution at 2.4 mA cm⁻² (corresponding to between 3.5 V — 3.7 V applied voltage) for 70 min. The Au-coated plate (1) and plain plates (2 — 10) were hot press assembled at 400 psig and 150 °C for 60 min, followed by a cooling phase of 120 min. Prepreg polymer sheets were used for adhesion between the PCB plates. The final assembled PCB cathode fractal flow-field had dimensions of 7.25 mm × 80 mm × 80 mm and is shown in Supplementary Information Fig. S1. Similarly, the cathode single-serpentine flow-field was Ni- and Au-coated, followed by a hot press assembly process, as above. The thickness of the cathode single-serpentine flow-field was similar to that of the fractal flow-field to ensure
identical insulation levels in both cells. The final assembled cathode single-serpentine flow-field with dimensions is also shown in Supplementary Information Fig. S1.

The anode flow-field had a single-serpentine square channel with an area of 1 mm² and a depth of 1 mm drilled on a 3 mm thick graphite plate. The dimensions of the final fabricated graphite-based anode single-serpentine flow-field were 3 mm x 80 mm x 80 mm. The anode current collector was developed from a PCB plate, having a 35 μm thick Cu coating and was coated with Ni and Au layers, using the aforementioned process.

2.3 MEA preparation

The MEA used for fuel cell testing had an effective area of 25 cm². The membrane used was Nafion® 212 (Dupont, USA) and the gas diffusion electrodes (GDEs) used were Hyplat Pt catalyst (HyPlat, South Africa) coated with a catalyst loading of 0.4 mgPt cm⁻². The gas diffusion layer (GDL) used was a carbon paper-based Freudenberg H23C9 with PTFE-treated microporous layer. The MEA components were hot pressed for 3 min at 400 psig and 150 °C, and the final assembled MEA had a thickness of approximately 500 μm. Two Tygaflor gaskets with a thickness of 250 μm were used to prevent gas leakage by sandwiching the MEA between them.

2.4 Fuel cell assembly

The fuel cell components and their order of assembly are shown in Fig. 2. The current-temperature mapping plate, S++ (S++ Simulation Services, Germany), was sandwiched between the anode flow-field and anode current collector. It is noteworthy that the mapping plate cannot be sandwiched at the cathode side, due to the electrically insulating nature of the PCB plates. Besides, the current collection on the cathode plate occurs via the surface. In
contrast, the mapping plate requires through-plane electrical conduction, which is possible with a conductive graphite plate on the anode side.

The S++ plate consisted of an array of $10 \times 10$ integrated shunt resistors, with each resistor having an area of $5 \times 5 \text{ mm}^2$, covering a total electrode area of $25 \text{ cm}^2$. The current measurements were made in through-plane mode. The surface temperature distribution measurement was carried out using a $5 \times 5$ array of temperature sensors. Current and temperature measurements were made simultaneously, and their respective sensors were located on the same sensor plate, made of PCB with Au-coated contact segments. The S++ plate was connected to a computer via a USB interface that provided live mapping (current/temperature) display and data recording. The reactants, air and $\text{H}_2$, are supplied to the cell as shown in Fig. 2. Air for both cathode fractal and cathode single-serpentine cells exited from the surface vertical flow paths (horizontal blue lines in Fig. 2) into the ambient atmosphere (Supplementary Information Fig. S1), while the $\text{H}_2$ outlet occurred from a manifold, as shown in Fig. 2. The corresponding air and $\text{H}_2$ flows are represented by blue and red lines, respectively, in Fig. 2. Compression of the cell components was provided using aluminium end-plates (cathode and anode), with a nut and bolt arrangement tightened to a torque of 1.4 N-m. The anode end-plate also acted as a heating device providing the necessary start-up temperature to the cells, which was 45 °C. The cell temperature was measured using a K-type thermocouple located within the anode flow-field plate.
Figure 2: Assembly outline and primary components used in the PEMFCs tested.

2.5 X-ray computed tomography (CT) scan

X-ray computed tomography (CT) scans were performed on the fractal cathode flow-field using a Nikon XT – H 225 instrument (Nikon Metrology, UK). The corresponding experimental setup, scan parameters, acquisition conditions, and the software utilised for data processing and image reconstruction are discussed in previous work [10].

2.6 Fuel cell testing

A Scribner 850e testing station (Scribner Associates NC, USA) was used to test the fuel cells. The testing station supplied the reactants at the desired temperature, reactant humidity (RH) and flowrates. The RH of the reactants was maintained by the test station by considering the cell temperature feedback and adjusting the corresponding reactant supply temperature. The fuel cells were supplied with hydrogen of 99.995% purity at a constant flow rate of 200 mL
min$^{-1}$, while the cathode airflow stoichiometry was maintained at 3.0. The cells were operated under ambient cooling conditions. After each experimental condition, the cells were flushed using nitrogen gas for 30 min on both cathode and anode regions of the cell to remove the excess water and reactants present inside it. Detailed experimental parameters are given in Table S1.

Electrochemical impedance spectroscopy (EIS) measurements were performed using a Gamry Reference 3000 and Gamry Reference 30k Booster (Gamry Instruments, USA). The frequency range for analysis was from 100 kHz to 0.1 Hz, with 10 points per decade and an AC modulation amplitude of 5% of the DC input signal.

3. Results and discussion

3.1 X-ray CT scan analysis

X-ray CT scan imaging of the fractal flow-field was performed to analyse the internal structure of the 5 generations of hierarchical fractal flow-field and the layer-wise cell assembly quality. The corresponding zx, xy and zy virtual slices are shown in Fig. 3a, 3b and 3c, respectively. The corresponding slice-by-slice 3D track through of the fractal flow-field in the zx, xy and zy directions can be accessed by clicking on Fig. 3. The dark regions, where the attenuation is lower, correspond to the flow channels, while the bright regions, where the attenuation is higher, correspond to the PCB material. The dark vertical lines observed in Fig. 3a correspond to the vertical flow paths, as shown in Fig. 2, which remove the excess reactants and product water from the cell. The five hierarchical fractal generations can be identified distinctly in Figs. 3b and 3c. Overall, the inter-channel arrangement, individual layer assembly and hierarchical flow paths are observed to be unobstructed, without any overlap, which establishes the PCB layer-wise assembly approach to be an effective method for developing fractal flow-fields.
Figure 3: Virtual slices in the (a) zx plane showing the 5th generation flow outlets and vertical flow paths, (b) xy plane and (c) zy plane showing the hierarchy extending over five generations for the studied fractal cathode flow-field.

3.2 Polarisation performance

The polarisation performance of the single-serpentine and fractal cells under different operating conditions are compared in Fig. 4. It can be observed that the electrochemical performance of the fractal cell is better than that of the single-serpentine cell in the range of reactant RH tested. For instance, at 0.6 V cell voltage, which is often considered to be an optimal operating point in the trade-off between efficiency and power [10], the corresponding current density output at 40%, 70% and 100% RH for the fractal cell is 0.65 A cm$^2$, 0.68 A cm$^2$ and 0.75 A cm$^2$, and for the single-serpentine cell is 0.54 A cm$^2$, 0.63 A cm$^2$ and 0.68 A cm$^2$, respectively. Furthermore, the limiting current densities at 40%, 70% and 100% RH for the fractal cell are 1.52 A cm$^2$, 1.59 A cm$^2$ and 1.66 A cm$^2$, compared to 1.44 A cm$^2$, 1.43 A cm$^2$ and 1.42 A cm$^2$ for the single-serpentine cell.
Figure 4: Polarisation curves for the fractal cell and single-serpentine cell at (a) 40% reactant RH conditions, (b) 70% reactant RH conditions and (c) 100% reactant RH conditions. Legend: $V_f$ – voltage of fractal cell, $V_s$ – voltage of single-serpentine cell, $P_f$ – power density of fractal cell and $P_s$ – power density of single-serpentine cell.

The overall enhanced performance of the fractal cell can be attributed to its fractal geometry-based cathode flow-field structure, where a hierarchically structured flow channel with equal hydraulic path lengths between inlet and outlet allows for the transition of airflow from a convection-dominated state at the inlet to a more diffusion-dominated state at the outlets, resulting in more uniform distribution of reactants over the MEA surface [7,8,10,40]. The effects of more uniform reactant distribution at the cathode of the fractal cell can be identified from its superior performance over the single-serpentine cell, especially in the high current density region between 1.2 A cm$^{-2}$ and 1.6 A cm$^{-2}$, where mass transport limitations occurring from reduced oxygen concentration in the cell are predominant, as shown in Fig. 4.
In addition to the more uniform reactant distribution, the presence of vertical flow paths (Supplementary Information Fig. S1) in the fractal cell act to regulate the excess water away from the MEA more effectively, avoiding excess stagnation of liquid water throughout the system [10]. As a result, while the performance of the single-serpentine cell does not vary appreciably with RH in the mass transport region, the performance of the fractal cell increases with reactant RH, which can be attributed to a reduced level of flooding and reactant (oxygen) starvation [10,41].

Table 1: Polarisation performances (current density) of fractal and single-serpentine cells at 0.6 V and 0.4 V cell voltages, and at 40%, 70% and 100% reactant RH conditions.

<table>
<thead>
<tr>
<th>PEMFC configuration</th>
<th>Cell voltage</th>
<th>Current density (A cm⁻²) at 40% RH</th>
<th>Current density (A cm⁻²) at 70% RH</th>
<th>Current density (A cm⁻²) at 100% RH</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fractal</td>
<td>0.6 V</td>
<td>0.65</td>
<td>0.68</td>
<td>0.75</td>
</tr>
<tr>
<td></td>
<td>0.4 V</td>
<td>1.25</td>
<td>1.35</td>
<td>1.37</td>
</tr>
<tr>
<td>Single-serpentine</td>
<td>0.6 V</td>
<td>0.54</td>
<td>0.63</td>
<td>0.68</td>
</tr>
<tr>
<td></td>
<td>0.4 V</td>
<td>1.17</td>
<td>1.23</td>
<td>1.24</td>
</tr>
</tbody>
</table>

3.3 Cell temperature analysis

The increase in cell temperature, recorded during the polarisation curve measurements in Fig. 4, is shown for each cell as a function of reactant RH in Fig. 5.
Simultaneous cell temperatures recorded during polarisation curve measurements, as in Fig. 4, using a single point thermocouple on the surface of the anode flow-field plate for the fractal cell and single-serpentine cell at (a) 40% reactant RH conditions, (b) 70% reactant RH conditions and (c) 100% reactant RH conditions. Legend: $V_f$ – voltage of fractal cell, $V_s$ – voltage of single-serpentine cell, $T_f$ – temperature of fractal cell and $T_s$ – temperature of single-serpentine cell.

The maximum cell temperature developed (at the limiting current density) in the fractal and single-serpentine cells, at 40%, 70% and 100% RH, are 70.3 °C, 72.2 °C and 74.6 °C and 64.9 °C, 65.5 °C and 67.1 °C, respectively. It can be observed that the fractal cell exhibited higher operating temperatures under all conditions, especially in the high current density region, compared to the single-serpentine cell. It is well established that the degree of liquid water saturation has a notable influence on the cell temperature developed [42].

Increased water retention in the cathode region reduces the current density generated and leads to a corresponding decrease in cell temperature and *vice versa* [5]. Thus, the lower cell
temperatures in the single-serpentine cell can be attributed to the presence of excess water saturation (flooding) inside the cell, despite operating at lower cell voltages compared to the fractal cell [43].

### 3.3 Electrochemical impedance spectroscopy (EIS) measurements

EIS was performed on the fractal and single-serpentine cells over a range of current densities and the corresponding Nyquist curves are given in Supplementary Information Fig. S2. The corresponding high frequency resistance (HFR) or ohmic resistance developed in the cells is given in Fig. 6(a). HFR is measured from the high frequency intercept of the Nyquist plot with the real axis [44]. It primarily reflects the membrane hydration state and its associated conductivity [45]. Fig. 6(a) shows that an increase in the reactant humidity from 40% RH to 70% RH and later to 100% RH results in a decrease in the HFR at the same current density, which can be attributed to the improved membrane conductivity with increasing RH. The HFR for each cell decreased with increasing current density, which can be attributed to improved membrane hydration as a result of the increased amount of water generated via the higher rate of the electrochemical reaction at the cathode. Overall, the observed trends in HFR are a well-established phenomenon that is common to both flow-fields and suggests that both cells were sufficiently hydrated throughout. The results also indicate that the level of hydration is higher in the fractal cell at all the RH levels tested [46,47]. Low frequency resistance (LFR), as shown in Fig. 6(b), is derived by fitting the Nyquist curves (Fig. S2) to an equivalent circuit given in Fig. S3 and corresponds to the total resistance developed in a cell [48].
Figure 6: (a) High frequency resistance (HFR or Ohmic) for fractal and single-serpentine cells at 40%, 70% and 100% RH reactant conditions and (b) low frequency resistance (LFR or total resistance) for fractal and single-serpentine cells at 40%, 70% and 100% RH reactant conditions. Legend - HFR$_{s}$ – HFR of single-serpentine cell, HFR$_{f}$ – HFR of fractal cell, LFR$_{s}$ – LFR of single-serpentine cell, LFR$_{f}$ – LFR of fractal cell.

For both cells, an initial decrease in LFR with increasing current density, as seen in Fig. 6(b), can be attributed to improved membrane hydration due to water generation at the cathode. This gradual reduction in LFR for both cells is observed between 0.2 A cm$^{-2}$ and 0.6 A cm$^{-2}$. At low and intermediate current densities, the LFR, at 40%, 70% and 100% RH, for the single-serpentine and fractal cells are similar, with, at 0.2 A cm$^{-2}$, 0.60 Ω cm$^2$, 0.57 Ω cm$^2$, 0.51 Ω cm$^2$ (single-serpentine) and 0.61 Ω cm$^2$, 0.55 Ω cm$^2$, 0.49 Ω cm$^2$ (fractal), and, at 0.6 A cm$^{-2}$, 0.39 Ω cm$^2$, 0.37 Ω cm$^2$, 0.36 Ω cm$^2$ (single-serpentine) and 0.36 Ω cm$^2$, 0.33 Ω cm$^2$, 0.32 Ω cm$^2$ (fractal). The LFR and HFR decreased between 0.2 A cm$^{-2}$ and 0.6 A cm$^{-2}$, as shown in Fig. 6(a).

However, when the current density increases to 0.8 A cm$^{-2}$ and further to 1 A cm$^{-2}$, a sudden rise in LFR for the single-serpentine cell is observed. This higher LFR for the single-serpentine flow-field at higher operating currents can be attributed to the increased charge- and mass transport related resistances, such as reactant starvation occurring from cell flooding [49]. The corresponding charge transfer resistances ($R_{ct}$) and mass transfer resistances ($R_{mt}$) for the
single-serpentine and fractal cells at 0.8 A cm\(^{-2}\) and 1 A cm\(^{-2}\) current densities are given in Table 2. In contrast, the LFR for the fractal cell continues to decrease throughout its operation, even in the high current density region, where \(R_{\text{ct}}\) and \(R_{\text{mt}}\) of the fractal cell are significantly lower than for the single-serpentine cell, which can be attributed to the flooding-free, diffusion-dominated oxygen mass transport [9].

Table 2: Charge transfer resistances (\(R_{\text{ct}}\)) and mass transfer resistances (\(R_{\text{mt}}\)) for single-serpentine cell and fractal cell at 0.8 A cm\(^{-2}\) and 1 A cm\(^{-2}\), for 40%, 70% and 100% RH reactant conditions.

<table>
<thead>
<tr>
<th></th>
<th>Single-serpentine</th>
<th>Fractal</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>40% RH</td>
<td>70% RH</td>
</tr>
<tr>
<td>(R_{\text{ct}}) (Ω cm(^{-2}))</td>
<td></td>
<td></td>
</tr>
<tr>
<td>@ 0.8 A cm(^{-2})</td>
<td>0.207</td>
<td>0.175</td>
</tr>
<tr>
<td>@ 1 A cm(^{-2})</td>
<td>0.194</td>
<td>0.172</td>
</tr>
<tr>
<td>(R_{\text{mt}}) (Ω cm(^{-2}))</td>
<td></td>
<td></td>
</tr>
<tr>
<td>@ 0.8 A cm(^{-2})</td>
<td>0.015</td>
<td>0.024</td>
</tr>
<tr>
<td>@ 1 A cm(^{-2})</td>
<td>0.016</td>
<td>0.024</td>
</tr>
</tbody>
</table>

3.5 Current distribution analysis

Current distribution maps at 40%, 70% and 100% reactant RH are presented in Fig. 7, where the maps (a-c), (g-i) and (m-o) correspond to the fractal cell and (d-f), (j-l) and (p-r)
Figure 7: Current distribution for fractal cell at (a-c) 40% RH, (g-i) 70% RH and (m-o) 100% RH, and for single-serpentine cell at (d-f) 40% RH, (j-l) 70% RH and (p-r) 100% RH. Cell voltage for (a, d, g, j, m, p) is 0.8 V, for (b, e, h, k, n, q) it is 0.6 V and for (c, f, i, l, o, r) it is 0.4 V.
correspond to the single-serpentine cell. The maps in Fig. 7 represent the local currents measured by the 10 × 10 shunt resistors (segments) on the current mapping plate, as shown in Fig. S4. A schematic representation of the cathode flow-fields, with respect to the current mapping segments, is given in Fig. S5.

At 0.8 V cell voltage, it can be observed from the maps in Fig. 7 that, irrespective of the reactant RH condition, a similar and uniform current distribution is measured in both single-serpentine and fractal cells. Furthermore, the mean of segment currents (MSC), an average of the overall segment currents measured, and the corresponding standard deviation (STDEV), representing variations in segment currents from the MSC, are given in Figs. 8 (a-b), and the corresponding MSC and STDEV at 0.8 V for both cell designs are similar at ≈ 0.025 A and ≈ 0.003 A, respectively.
Figure 8: Mean of segment currents (MSC) and its corresponding standard deviation (STDEV) for fuel cells with (a) a single-serpentine flow-field (S) and (b) a fractal flow-field (F), at 40%, 70% and 100% reactant RH, and (c) normalised STDEV with respect to the MSC for the same.

In addition, the normalised STDEV (STDEV/MSC) for both cells are almost the same, indicating similar levels of dispersion of segment current around the MSC, as shown in Fig. 8 (c). This is supported by the segment current distribution about the MSC at 0.8 V, as shown in Fig. 9, which indicates uniform current development across all 100 segments (segment-wise representation on the current maps is shown in Fig. S5). Furthermore, Fig. S6 represents the uniformity of the current distribution as a function of cell voltage, but with different current scales adjusted for each cell voltage and the segment wise current distribution across the 10 x 10 segments.
Figure 9: Segment current distribution at (a) 0.8 V cell voltage, (b) 0.6 V cell voltage and (c) 0.4 V cell voltage. Legend: MSC – mean segment current, S – single-serpentine, F – fractal.

These imply that the overall current distribution is uniform across segments in both cells at low operating currents [23,50]. Such homogeneity in the current distribution, at higher cell voltages, can be attributed to the negligible diffusion limitations, since the intrinsic rate of reaction and current are low, with minimal impact from water dynamics and reactant.
concentration [22,37]. Here, the observed similarities in current distribution between the cells corroborate with the polarisation performances observed in Fig. 4, where, at 0.8 V, each cell delivered an output of ≈ 0.1 A cm$^{-2}$ = (100 × 0.025 A)/25 cm$^2$.

However, at lower cell voltages, the current distribution is inhomogeneous, as seen in Fig. 7, which can be attributed to diffusion limitations at the corresponding higher average currents. Prominent low-current regions, seen as deep blue patches in Fig. 7, developed to a larger extent in the single-serpentine cell. Comparatively, the fractal cell developed a more uniform current distribution. Besides, the fractal cell is observed to have a higher MSC and lower STDEV compared to the single-serpentine cell, as shown in Figs. 8 (a-b). The corresponding normalised STDEV in Fig. 8 (c) is also lower for the fractal cell, confirming lower levels of dispersion of segment currents around the MSC. However, the greater extent of low-current regions in a single-serpentine cell likely results from relatively excessive reactant starvation in these regions, due to localised flooding or excess water retention [51–53]. This is supported by the higher levels of dispersion of segment currents around the MSC, as established by the normalised STDEV shown Fig. 8 (c). The corresponding segment current distribution (100 segments) around the MSC, at 0.6 V and 0.4 V cell potentials, is given in Fig. 9, which highlights the development of low and high segment currents in each cell. However, the number of such segments varies, depending on the cathode flow-field type used (note again that a single-serpentine anode flow-field was used in both cases). In particular, the number of low current segments developed in a fractal cell is much lower than that in a single-serpentine cell under any condition, consistent with a lower normalised STDEV in Fig. 8 (c). For instance, ≈ 8 – 10 segments in a fractal cell and ≈ 18 – 20 segments in a single-serpentine cell develop a current
less than 0.1 A at 0.6 V, and 0.2 A at 0.4 V, respectively (the currents here correspond to the 
value for MSC-STDEV for a single-serpentine cell at the corresponding voltage). Overall, the observed better performance with more homogeneous current distribution for 
the fractal cell can be attributed to the uniform reactant and water distribution occurring 
from the fractal flow-field structure. In addition, the better performance of the fractal cell 
identified here corroborates the corresponding polarisation performances, given in Table 1.

3.6 Surface temperature distribution analysis

The corresponding anode surface temperature distribution, indicative of the local internal cell 
temperatures in the fractal and the single-serpentine cells, is given in Fig. 10. Here, the 
hydration conditions generated inside the cell and their influence on the corresponding 
surface temperature are explored. The surface temperatures measured at the central 
segment of the mapping plate (at the centre of the anode surface) for the fractal cell at 0.6 V 
are 53.4 °C (40% RH), 55 °C (70% RH) and 56.1 °C (100% RH), and at 0.4 V are 64.6 °C (40% 
RH), 66.1 °C (70% RH) and 71.9 °C (100% RH). Corresponding central segment temperatures 
for the single-serpentine cell at 0.6 V are 51.4 °C (40% RH), 52.8 °C (70% RH) and 54.5 °C 
(100% RH) and at 0.4 V are 62.4 °C (40% RH), 63.6 °C (70% RH) and 65.4 °C (100% RH), 
respectively. It is clear that the fractal cell developed much higher surface temperatures in 
the central segment of the anode plate compared to the single-serpentine cell, irrespective 
of reactant RH and cell operating voltage, corroborating the cell temperatures measured 
using a thermocouple during polarisation curve measurements, as given in Fig. 5.
Figure 10: Temperature distribution for fractal cell at (a-c) 40% RH, (g-i) 70% RH and (m-o) 100% RH, and for single-serpentine cell at (d-f) 40% RH, (j-l) 70% RH and (p-r) 100% RH. Cell voltage for (a, d, g, j, m, p) is 0.8 V, for (b, e, h, k, n, q) it is 0.6 V and for (c, f, i, l, o, r) it is 0.4 V.
Besides, the surface temperatures are 4 – 5 °C lower than the cell temperatures measured using the thermocouple, across all operating conditions. This can be attributed to the proximity of the thermocouple to the MEA region, in addition to the heat loss occurring to the ambient surroundings.

With continuous heat dissipation to the surroundings at the edges of the cell, the temperature is higher in the cell centre and lower at the edges, as seen in Fig. 10 [38,54,55]. The differences between the centre and edge surface temperatures are similar for both cells: -0.2-3 °C, -1-2 °C at 0.6 V and -3-4 °C at 0.4 V, irrespective of the reactant conditions, indicating that there is a consistent temperature gradient or heat distribution on the cell surface for a given current density under the range of conditions studied.

The lower surface temperatures measured in the single-serpentine cell can be attributed to the reduced rate of heat dissipation compared to the fractal cell. Another contributing factor is the localised flooding occurring in the single-serpentine cell, especially on the GDL surface, which reduces the effective catalyst area available for the electrochemical reactions to occur, and results in decreased current density and associated generated heat [56].

4. Conclusions

An electro-thermal mapping approach has been applied to derive original insights into the reasons for improved performance of a cathode fractal flow-field compared to a conventional serpentine flow-field for a PEMFC. The study establishes a relationship between cell performance and the surface distribution of current and temperature. It also provides an understanding of localised phenomena, such as flooding occurring because of the flow-field configuration.
X-ray CT scan analysis demonstrates the layer-wise PCB-based assembly as an effective approach for developing the fractal flow-fields. The fractal fuel cell delivered better performance, especially in the high current density region, compared to the single-serpentine cell, over a range of reactant RH and operating conditions. The enhanced performance of the fractal cell is a result of higher cell temperatures developed in the fractal cell compared to the single-serpentine cell. Uniform and stable Ohmic resistances, over the polarisation range, are measured for both cells, indicating well-hydrated membrane conditions throughout operation. In addition, electrochemical impedance spectroscopy measurements are consistent with stable and flooded operating conditions in the fractal and the single-serpentine cells, respectively.

Relatively homogeneous current distribution is observed in the fractal cell as a result of more uniform reactant and water distribution, while a more non-uniform current distribution is developed in the single-serpentine cell as a result of flooding conditions. The superior performance of the fractal cell is reflected by the segment currents measured, which are ≈ 10-15% higher than those measured in the single-serpentine cell. This is consistent with the higher surface temperature distribution observed for the fractal cell as a result of the higher current density.

Finally, this study further asserts, via electrothermal metrology, the water and thermal management benefits in fuel cells when using a nature-inspired approach, compared to conventional fuel cells.
Acknowledgements

The authors would like to acknowledge funding from EPSRC “Frontier Engineering” (EP/K038656/1) and “Frontier Engineering: Progression” (EP/S03305X/1) Awards to the Centre for Nature-Inspired Engineering (CNIE), as well as other funding from the EPSRC (EP/L015277/1, EP/P009050/1, EP/M014371/1, EP/M009394/1, EP/M023508/1, EP/L015749/1, EP/N022971/1) for supporting fuel cell research in the Electrochemical Innovation Lab (EIL). We also thank the Department of Chemical Engineering at UCL, and the National Measurement System of the UK Department of Business, Energy and Industrial Strategy for supporting this work.

The authors would like to acknowledge Lara Rasha and Yunsong Wu, PhD students from the Department of Chemical Engineering at UCL, for their assistance in sourcing the equipment and supplying the interfacing code used to obtain the data.

5. References


[37] I. Alaefour, G. Karimi, K. Jiao, X. Li, Measurement of current distribution in a proton exchange membrane fuel cell with various flow arrangements - A parametric study,


[50] D.J.L. Brett, S. Atkins, N.P. Brandon, V. Vesovic, N. Vasileiadis, A. Kucernak, Localized impedance measurements along a single channel of a solid polymer fuel cell,


