Experimental report

24/06/2022

Proposal: **UGA-130** **Council:** 4/2021

Title: In operando neutron tomography of polymer electrolyte fuel cell wateraccumulation

Research area:

This proposal is a new proposal

Main proposer: **Jennifer HACK**

Experimental team: Alessandro TENGATTINI
Ralf ZIESCHE
Theo SUTER
Jennifer HACK
Matilda FRANSSON

Local contacts: Lukas HELFEN
Alessandro TENGATTINI

Samples: Water
Hydrogren (N4.5 grade (99.998%))
Nitrogen and air
Membrane electrode assemblies (MEAs) consisting of solid Nafion membrane between two solid platinum/carbon electrodes (2cm2) housed in a gold-coated aluminium cell

Instrument	Requested days	Allocated days	From	To
NEXT	5	5	09/07/2021	14/07/2021

Abstract:

1 Background

Operando neutron computed tomography (NCT) is an emerging technique for studying water evolution in polymer electrolyte fuel cells (PEFCs), with our recent work showing the first example of high-speed NCT for studying water in the channels of a single serpentine (SS) flow field over the first minutes of fuel cell operation (10 min current holds) [1]. Previous studies had relied either on radiography [2, 3], or had used in-situ [4,5] CT (with scan times lasting hours) for observing water.

In this work, the high-flux beam available on the NeXT (D50) beamline of the Institute Laue Langevin (ILL) has allowed for improved image quality, whilst maintaining high-speed imaging with <40 s/tomography. Since the current holds in our previous work [1] were limited to 10 min/hold, questions arose about the water evolution during 'steady-state' operation, *i.e.* over a 1 h current hold. Furthermore, the effects of different flow channels on water distribution and removal were also of interest.

2 Experiment details

A total of 120 hrs of beamtime were used for this experiment (since experiments 005-014-Maier and 005-016-Hack were combined after a delay in scheduling due to the COVID-19 pandemic). In collaboration with the imaging group from Helmholtz Zentrum Berlin (HZB), the NeXT instrument was prepared for high-speed imaging and optimised with respect to a maximum of neutron flux and appropriate spatial resolution. Scans were collected with 40 Hz resulting in 1440 projections over 360°, equalling 36 s/tomogram. The pixel size was around 64 µm. Reconstructions were done by using the ASTRA Toolbox in a python script. All projections were dark-field corrected, normalised and outlies, bright spots from neutrons which directly hit the camera chip, were filtered. Finally, the data were sequential reconstructed with 180° steps between each NCT. After reconstruction, the cathode and anode flow channels and the MEA area were defined and the water volume calculated from the grey values as described in detail in [1].

Three flow field designs were studied, namely single serpentine (SS), double serpentine (DS) and parallel (PAR). The active area of each cell was 2 cm^2, though there is scope to increase this in future experiments to optimise the field of view. Slip rings enabling both gas flow and electrical connection were attached to the bottom and top of the cell to allow for continuous rotation during imaging. Unfortunately, due to COVID-19 and the remote nature of the experiment, it was not possible to heat the cells as planned, and this should be done in future experiments. Experiments were carried out with 100 mL min^{-1} flow rate on the cathode and two different gas flow rates on the anode, namely 20 mL min^{-1} and 50 mL min^{-1}, to understand the effect of increased flow rate on water evolution. For each cell, after collecting three initial polarisation curves, 1 hr current density holds between 100 – 600 mA cm^{-2} were collected in 100 mA cm^{-2} intervals at 20 mL min^{-1} flow rate, then between 600 – 100 mA cm^{-2} at 50 mL min^{-1} flow rate. Some cells did not reach 600 mA cm^{-2}, due to performance limitations.

In addition to the beamline scientists, one experimental team member from UCL was on site, but other team members were working remotely and the experiment largely ran remotely. Set-up of the fuel cell and camera system took 24 hrs, due in-part to the mainly-remote nature of the experiment, as well as the complex set-up. Following this, each set (SS, DS, and PAR) took ~24 hrs to complete (factoring in time for cell de-activation at the end of each experiment). SS experiments were repeated to use the final 24 hrs of experimental time.

3 Preliminary results and discussion

Polarisation and power density curves showing the performance of the three cells are shown in Figure 1. Whilst the SS and DS cells have similar performance, with current densities of 657 and 654 mA cm^{-2}, respectively, at 0.3 V, the PAR cell has a lower current density of 578 mA cm^{-2} at 0.3 V. This is also shown by the power density curves, with the SS and DS having peak power densities of 207 and 205 mW cm^{-2}, respectively, and the PAR cell having a peak power density of 184 mW cm^{-2}. This disparity in performance suggests that flooding is more significant in the PAR cell, which lowers the performance at high current densities, than for the SS or DS cells.

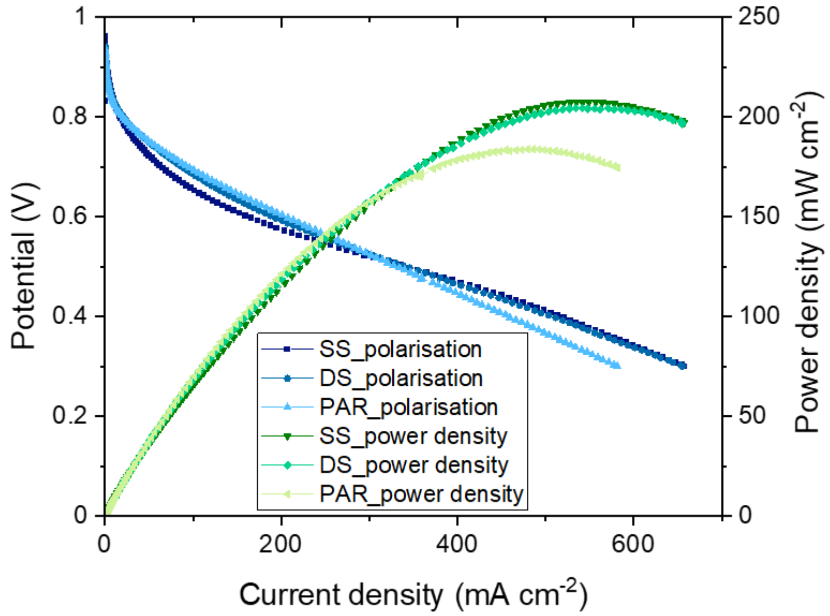

Figure 1 Comparison of fuel cell performance of the cells with three different flow fields, namely single serpentine (SS), double serpentine (DS) and parallel (PAR). Polarisation curves are shown in shades of blue and power density curves are shown in green/

To date, most of the CT datasets have been reconstructed and an example of some preliminary analysis of the results is shown in Figure 2 (SS flow field, 1 h hold @ 400 mA cm^{-2}, 50 mL min^{-1} flow rate). As can be seen for the graph of the water volume (Figure 2a), the volume of water steadily increases in the cathode, until it reaches a plateau at ~3.5 mm^3 between 750 and 1116 s. This can be seen in Figure 2 b and c, (3D volume and 2D orthoslices, respectively, at 1008 s), where there are water droplets in the bends of the channel at the top of the cell. After 1116 s, the water volume begins to decrease, suggesting that the rate of water removal from the cell is faster than the rate of water production. After 1728 s (~30 mins), the water volume reaches a constant value of ~0.5 mm^3, which suggests than an equilibrium between water formation and water removal in the gas stream has been reached. There will also be an increase in cell temperature as a result of the exothermic reaction, which could contribute to water removal. The water volume in the anode is mostly constant at ~0.3 mm^3, which is expected given that water production occurs on the cathode side. It also means there is no back-diffusion of water, which indicates a good cell setup. Finally, after an initial increase, the water volume in the MEA reaches a constant value of around 1 mm^3.

In continuing the data analysis of the CT datasets, the evolution of water across the different current holds will be analysed, as well as understanding the effect of anode flow rate on the water volume in the cell.

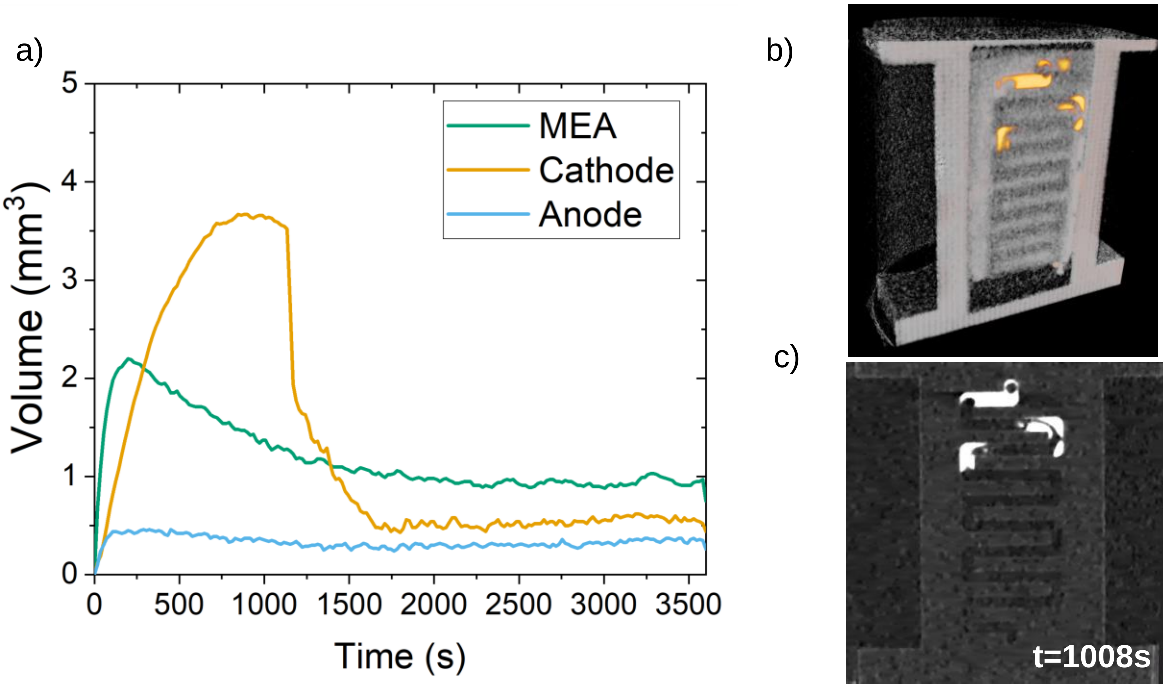

Figure 2 a) Volume of water for a single serpentine cell in the MEA (green), cathode flow field (orange) and anode flow field (blue) measured from the tomograms over the 1 hr current hold at 400 mA cm^{-2}; b) volume rendering of the SS cell at 1008 s with cathode water shown in orange and c) orthoslice of the cathode flow field.

4 Conclusions and future work

High-speed 4D tomography of fuel cells was carried out for three different flow fields (SS, DS and PAR), over a 1 hour current hold at various current densities and flow rates. This allowed for the evolution of water in the flow channels to be studied over a longer duration (*i.e.* steady-state) than were done in previous work [1]. Unfortunately, due to the COVID-19 pandemic, it was not possible to operate cells under heating and this should be done in future work. In addition, the grade of aluminium/gold-coating has meant the cells have taken a long time to become de-activated, thus a different grade should be used in future. Nonetheless, the experiment was deemed a success and once the continuing data analysis is complete, it is expected that a paper will be published in the coming year.

5 References

[1] Ziesche, R.F., Hack, J., Rasha, L., Maier, M., Tan, C., Heenan, T.M., Markötter, H., Kardjilov, N., Manke, I., Kockelmann, W. and Brett, D.J., 2022, *Nature communications*, 13(1), 1-11.

[2] Wu, Y., Cho, J.I.S., Neville, T.P., Meyer, Q., Ziesche, R., Boillat, P., Cochet, M., Shearing, P.R. and Brett, D.J.L., 2018, *Journal of Power Sources*, 399, 254-263.

[3] Kulkarni, N., Cho, J.I., Rasha, L., Owen, R.E., Wu, Y., Ziesche, R., Hack, J., Neville, T., Whiteley, M., Kardjilov, N. and Markötter, H., 2019, *Journal of Power Sources*, 439, 227074.

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