Experimental report

06/09/2018

Proposal: TEST-2899 **Council:** 4/2018

Title: Characterisation of hydrogen fuel cells by neutron tomography on D50-Tomo

Research area:

This proposal is a new proposal

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Samples: C + Pt + PTFE (CF2)n

Instrument	Requested days	Allocated days	From	To
D50 T	2	2	21/06/2018	23/06/2018

Abstract:

Experimental Report (Cycle 182)

TEST -2899: Characterisation of hydrogen fuel cells by neutron tomography on D50-Tomo

Scientific Context

The PEMFC consists of multiple layers clamped between two flow fields as seen in Figure 1. The gas diffusion layer (GDL), a sheet of porous carbon composite, facilitates the transports of reactants and product between the flow field and the catalyst layer (CL). The electrochemical reactions take place in CL that contains ionomer as well as catalyst particles embedded in nanocarbon. The heart of the fuel cell is the membrane that permeates protons (H^+) from anode to cathode, yet neither electrons nor reactants. The most commonly used membrane is a sulfonated tetrafluoroethylene film which was first developed by DuPont (Nafion®) in 1960s. It Is necessary to develop alternative membranes that delivers the comparable performance to Nafion®, but at the reduced price. The proton conductivities of both Nafion® and alternative membranes increase with the level of hydration. The change in ohmic resistances (measured at high frequency) represents the change in the membrane hydration, given that the resistances of the other fuel cell components remain constant during the operation. The hydration can be provided by pre-humidifying reactant gases and/or generating current. By the high-resolution neutron imaging at D50 of the Insitut Laue-Langevin, we quantified water content in the fuel cells containing Nafion and alternative membranes (N212 and IN1515, respectively).

Data Treatment

The internal components of the fuel cell were successfully visualized at D50 beamline as seen in Figure 1 (a). The spatial and temporal resolutions were 8.5 µm/pixel and 0.1 frames per second, respectively. During image acquisition, the fuel cells were operated at 80°C at current densities ranging from 0.05 to 1.6 A/cm^2, for which the corresponding voltage responses and ohmic resistances are shown in Figure 2. The two fuel cells (i.e. one with N212 and the other with IN1515) were operated under the identical operating conditions. The Beer-Lambert law was applied to the raw radiographs to identify the water quantity in the fuel cell components. The images of the fuel cell under current generation (water production) was normalized by one during the open circuit voltage state (no water production). Figure 3 describes the average water content in the membrane over time (frames) for two fuel cells. The data was processed by using ImageJ software.

The Use of Beamtime

For this experiment, 48 hrs of beamtime was allocated. The beamline was fully functional without any downtime. The initial setup and alignment of the sample required substantial maneuvers and took about 12 hours. First, we tested fuel cells under dry, 50, 80, and 100 humidity levels without current to compare the level of membrane hydration, each condition lasted for 0.5-1.0 hr with a stabilisation period (15 min). The, the fuel cells were operated with current at 80 and 100% RH under various conditions (flow rates and pressure). For the change of the fuel cell on the sample stage, 3-4 hours of cool down period was required to ensure that the radiation level was under the safety threshold. We encountered unexpected problem with our fuel cell test bench, which was resolved by replacing it with a backup test bench. Despite the unexpected issues, we acquired valuable data for the investigation of alternative membrane.

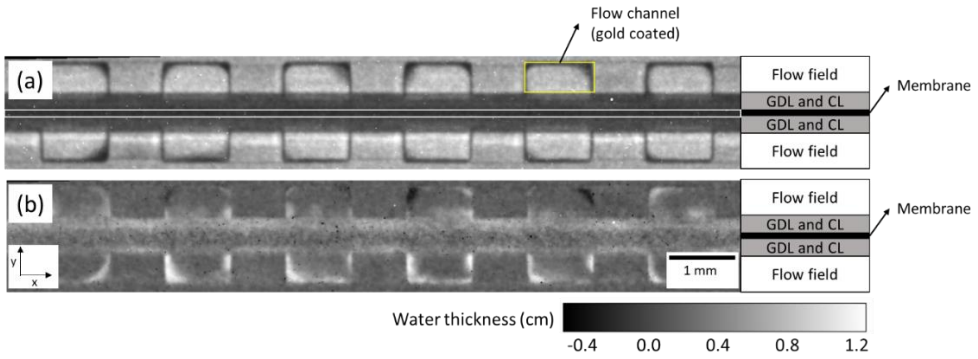

Figure 1: (a) an example raw radiograph, and (b) an example processed radiograph (water quantified in thickness).

Preliminary Results and Discussion

The fuel cell with Nafion® membrane exhibited superior performance to that of the fuel cell with alternative membrane at current densities higher than 0.8 A/cm^2. This current density approximately corresponds to frame number 200 in Figure 3. This performance difference between two fuel cells is strongly attributed to the water content in the membrane.

For N212 membrane, the water content in Figure 3 reached a plateau before 0.8 A/cm^2, and after then it manifested a sharp increase. The water content, once again, established the second plateau. This behaviour could be explained by Schroeder's paradox, which describes the difference between saturations equilibrated by gaseous and liquid phases. In other words, the ionomer retains a higher number of water molecules when it is hydrated by liquid water than by water vapour. At the current densities lower than 0.8 A/cm^2, the product water existed in vapor phase due to the heat produced in the CL. At 0.8 A/cm^2, we observed the onset of the liquid water accumulation in the CL and the GDL due to the increased water production. This observation supports the presence of Schroeder's paradox in our experiment.

On the other hands, the water content in IN1515 membrane showed an interesting trend at current densities above 0.8 A/cm^2. The water content started to decrease at 0.8 A/cm^2, but it began to recover at 1.2 A/cm^2. In general, the water content in the membrane is directly correlated to the ohmic resistance of the fuel cell. However, Figure 2 shows that the increase in ohmic resistance did not exhibit any relationship with the water content. We hypothesize that this ohmic resistance increase was attributed to the loss of an ability to retain water in the polymer matrix of the alternative membrane at 80°C. The wrinkles (or surface unevenness) in the membrane was visually observed when the fuel cell was disassembled after the experiment. We believe that the wrinkles may have affect on the quantity of water in the membrane, particularly liquid water accumulation on the membrane surface vs. water uptake by the membrane. We will investigate the performance and the properties of the alternative membrane at high temperature to provide an insight into the observed phenomena.

Conclusion

In this work, we successfully visualized water content across the fuel cell component in through-plane direction. Two types of membrane (conventional Nafion® and alternative) were investigated. The

neutron radiographs provided significant insight into establishing a link between physical phenomena and electrochemical measurements of the fuel cell.

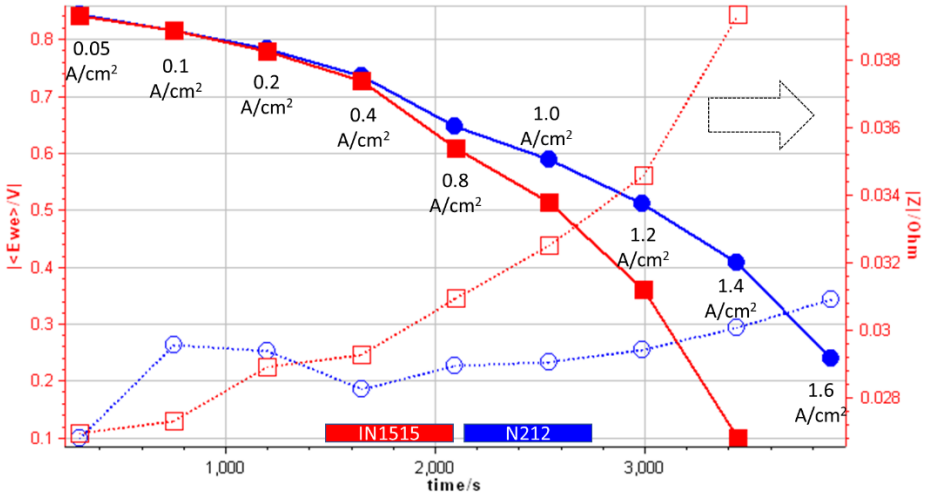

Figure 2: Voltage responses of fuel cells containing N212 (solid blue) and IN1515 (solid red) at current densities ranging from 0.05 to 1.6 A/cm^2. The corresponding ohmic resistances are displayed in dotted lines and empty markers.

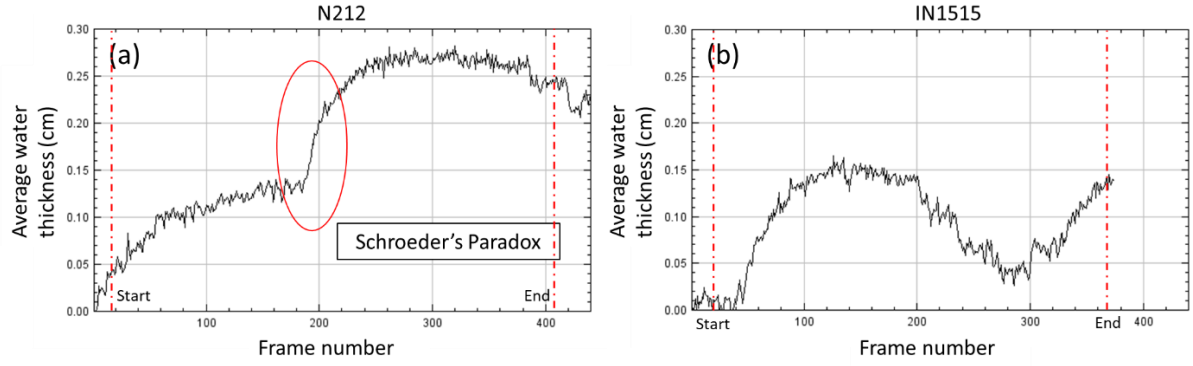

Figure 3: Average water content in the membrane of (a) N212 Nafion® membrane, and (b) IN1515 alternative membrane. The highlighted region in (a) demonstrates the effect of Schroeder's paradox on the level of membrane hydration.