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Water management in a planar air-breathing fuel cell array using operando neutron imaging

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HIGHLIGHTS

- An original micro fuel cell design with planar interconnections is presented.
- The huge sensitivity of breathing fuel cells to hydration mechanisms is detailed.
- The importance of interconnects and packaging elements is highlighted.
- The water nucleation pattern is found strongly influenced by convective flows.

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ABSTRACT

Operando Neutron imaging is used for the investigation of a planar air-breathing array comprising multiple cells in series. The fuel cell demonstrates a stable power density level of 150 mW/cm^2. Water distribution and quantification is carried out at different operating points. Drying at high current density is observed and correlated to self-heating and natural convection. Working in dead-end mode, water accumulation at lower current density is largely observed on the anode side. However, flooding mechanisms are found to begin with water condensation on the cathode side, leading to back-diffusion and anodic flooding. Specific in-plane and through-plane water distribution is observed and linked to the planar array design.

1. Introduction

The demand for portable energy sources has dramatically increased with the development of high energy consuming mobile devices, such as smartphones and notebook computers. The autonomy of these products is of great concern, and Micro-fuel cells, especially Proton Exchange Membrane Fuel Cell (PEMFC) have been investigated as an alternative to batteries.

In order to fit the current and voltage requirements of the majority of portable electronic devices, planar arrays, comprising multiple adjacent cells connected in series, have been developed [1–4].

Open cathode systems, also known as air-breathing fuel cells, offer the possibility to simplify the operational mode and hence to develop enlightened systems. However, despite similarities in behavior and materials, the breathing mode differs from the classical stack architecture in some crucial points, such as the power densities level, which is between 100 and 200 mW/cm^2 [5,6], and the absence of control in temperature and humidity level of the reactants. The performance of air breathing PEM fuel cells is consequently known as strongly limited by the free convection on the cathode side. The low heat and mass transfer coefficients on the cathode side control the heat dissipation, the oxygen supply and the removal of water [7]. The exponential increase of the saturation pressure of water vapor leads to a high sensitivity of the fuel cell performances to the temperature [8]. The design strategies often used to control the performances of breathing fuel cells tend to evacuate heat and retain water on the cathode side [1,9–11]. These strategies however make the fuel cell very sensitive to flooding issues, especially when the yield is increased or when the fuel cell

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works in a low temperature environment. Understanding the behavior of these systems with respect to water management is consequently crucial for their development.

Water distribution in PEMFC is a great concern since the first study in 1997 by Büchi et al. [12]. Depending on the conditions (gases humidity [13,14], stoichiometry [15], flow field design [16,17]), cathode or anode flooding is reported.

The nucleation of liquid water has been extensively studied in conventional fuel cells (where gas distribution and current collection is provided by bipolar plates), exhibiting power densities superior to 500 mW/cm^2. X-ray scattering, neutron imaging and scattering as well as Raman spectroscopy have been developed for in-situ and operando studies of water distribution [18]. Several studies [17–21] proved the efficiency of neutron radiography to visualize and quantify water in an operating PEMFC. However, few studies are focused on self-breathing PEMFC. Experimental studies deal mainly with the direct optical observation of water accumulation. Lee et al. [14] and Karst et al. [10] studied the water nucleation in the anodic compartment with dead-end configuration by using a transparent cell for optical observation of water droplets nucleation. Useful information results from these observations but the water visualization is partial, and the replacement of the anodic chamber by a transparent media for observation can affect the fuel cell behavior.

In the present paper, operando neutron imaging is used to visualize water patterns on a multiple cells planar breathing Printed Circuit Board (PCB) based fuel cell. The water patterns at different yields are discussed, in correlation with the performance of the fuel cell, the thermal gradients and the power density levels. The impact of connecting the cells in series in the plane of the fuel cell is examined with respect to water and performance distribution.

2. Experimental setup

2.1. Fuel cell design

The studied fuel cell is composed of ten in plane interconnected cells, with a breathing cathode (see Fig. 1). A Printed circuit board (PCB) support, composed of FR4 material (FR4 - Flame Resistant 4 is a composite material composed of a fiberglass cloth with an epoxy resin binder) and Gold/nickel coated copper traces, ensures the current collection and cell interconnections. Far from the PCB ribs, the current collection is achieved by a porous gold layer. More precisely, the Nafion® XL membrane is coated on both sides with a catalyst ink, composed of Pt/C (70 wt %), 5% Nafion® dispersion, solvent being a mixture of water and isopropanol. The copper traces are approximately 100 μm thick and covered with Ni/Au thin layers. Hydrogen is supplied via a laboratory-designed multi stage anodic chamber, providing a homogeneous gas feed.

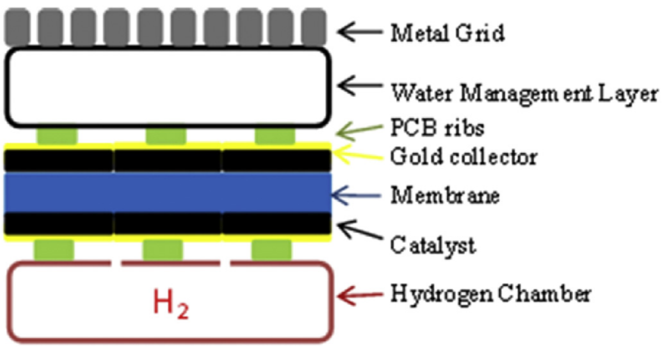

Fig. 1. Scheme of the planar fuel cell with the covering layer - cross-section view of the fuel cell core.

During the experiment, the fuel cell stands vertically and is operated in dead-end mode (there is no hydrogen exhaust). Hydrogen is delivered by a Sodium Borohydride cartridge, which is able to deliver 24L of hydrogen. A desiccant tube is used to provide dry hydrogen to the fuel cell, and a pressure limiter set to 0.4 bar (relative pressure, anode side). The hydrogen is being synthetized by $NaBH_4$ hydrolysis in the request of the fuel cell, the total amount of hydrogen is thus limited to the dead volume (tubing, anode chamber …), making the experiment very safe. On the cathode side, insulating water management layer (WML) is sandwiched between the electrode and the metal grid of the test fixture. The WML consists in a 190 μm thick microporous hydrophobic plastic material which aims essentially at retaining water (porous PTFE membrane). The opening ratio of the test fixture is 15% (see Fig. 1). The role of this grid is mainly to ensure a constant pressure on the cathode side and to dissipate heat (it has no electrical contribution).

2.2. Beam parameters and image processing

The neutron imaging experiments were carried out on G3bis-Imagine beamline at Orphée Reactor in Saclay (Leon Brillouin Laboratory, France), that delivers a flux of 2.10^7 neutrons/s/cm^2 [22]. Beamline settings were chosen to minimize noise and to offer a sharp contrast, rather than searching a high spatial definition. Therefore, we used the following configuration:

Images were acquired thanks to a 100 μm scintillator and an Andor Neo sCMOS camera. The overall spatial resolution can be estimated to 200 μm. Pictures of the open beam and of the background noise (in absence of neutron beam) were preliminary acquired. For an accurate measurement of thin layers of water, the highest possible signal-to-noise ratio is needed. To evaluate the contribution of the scattered neutron background, we recorded images of the fuel cell + test fixture, with neutron absorbing elements placed along the x and y axis of the fuel cell. By measuring the neutron counts behind these elements, it was possible to build a "scattering image" which was further subtracted to all pictures [23]. ImageJ was used as processing software. The scattering intensity was found in the range of 10%–15%.

Quantitative determination of water thickness can be achieved using Beer Lambert's law:

$$I_t = I_0 e^{-\Sigma\delta} \tag{1}$$

Where:

I_t is the transmitted intensity,
I_0 the incident intensity of the neutron beam,
δ the thickness of absorbing material (mm)
Σ the linear attenuation coefficient (mm^{-1})

To establish the relation between attenuation and water thickness, a 1 mm thick water scale was used as reference. The ratio between the two transmissions (with/without water) is called relative neutron transmission (Tr) [23].

The relative transmission coefficient T_r correlates the water thickness with the linear attenuation coefficient such as:

Acquisition time	30 s
Aperture diameter	16 mm
Source-detector distance	4 m
Object-detector distance	4 cm