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High resolution neutron imaging for pulsed and constant load operation of passive self-breathing polymer electrolyte fuel cells

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ABSTRACT

Small scale passive self-breathing micro fuel cells are an interesting alternative approach to power small integrated portable systems. The water management of these cells is greatly affected by the cell design – especially at the cathode side. Different cathodic current collector designs with different opening ratios are studied considering the impact onto the water distribution by using in situ high resolution neutron imaging. The impact of pulsed and constant load operation is analyzed as well as the impact of different environmental conditions. Several key observations could be made concerning water management in such cells, including the absence of water accumulation in the anode channels as well as the absence of excessive flooding of the cathode gas diffusion layer (GDL), even in designs implying lateral transport of liquid water over relatively large distances (up to 5 mm).

1. Introduction

The development and increasing use of portable autonomous systems leads to a strong demand for autarkic power sources [1]. Although there are several approaches on collecting energy from the environment by energy harvesting [1–6], these solutions are limited to a specific environment. Thus in general, there is a need for integrated energy storage. Providing potentially high energy densities and efficiencies, fuel cells are an interesting approach and an alternative to batteries for small scale power supplies. In order to obtain high energy and power densities, the whole system needs to be miniaturized. However, peripheral components, such as valves, pumps and humidifiers can only be scaled down in a limited way. As a consequence, it is necessary to forego these components and design predominantly passive systems. In our approach, an entirely passive fuel cell is developed that operates on dry hydrogen provided by a galvanic hydrogen generation cell [7] and uses oxygen from the surrounding air at the open self-breathing cathode. In this setup, the design of the cell components is of great importance, especially in terms of oxygen supply and water management.

Apart from providing a path for oxygen transport, the cathodic gas diffusion layer (GDL) and micro structured current collector also act as key components to influence the water transport towards the environment. Thus, these components have a strong impact on the water management and humidification of the polymer electrolyte, which relies on self-humidification in the passive cell. The significance of the openings in the cathodic current collector on the cell performance was shown in [8,9]. The strong gain in cell power for structures with a smaller opening ratio is believed to be mainly originating from improved self-humidification of the polymer electrolyte.

The development of passive self-breathing micro fuel cells predominantly focuses on technological concepts [8–21]. Jeong et al. [16] observed an increase of ohmic losses and activation losses with an increased cathodic opening ratio resulting from an increased evaporation of product water. Water management in passive self-breathing cells was also addressed by Karst et al. [20], who studied the impact of the opening ratio of an external cathodic cover for an opening ratio of 5%, 40%, and 100%and observed a significant performance improvement for the low opening ratio under dry conditions (30 °C, 10% relative humidity). Fabian et al. [21] added an electrically conductive hydrophilic wick in between GDL and current collector, resulting in a successful operation even under

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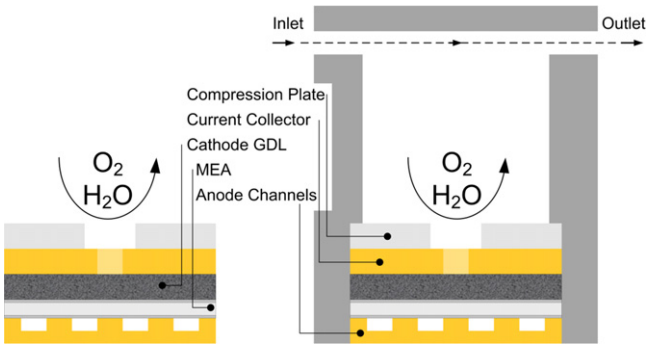

Fig. 1. Cell design principle and setup of closed cell for radiography measurements.

conditions of high humidity for various current densities with an anode dead-end configuration.

Neutron radiography is a well-known approach for water visualization in fuel cells, and has been used in numerous experimental studies [22–38]. Among these studies, only a few have addressed the effect of passive operating configurations, by studying fuel cells with a dead-end operated anode [37,38]. The visualization of water accumulation in an anode with dead-end configuration was also reported by Lee et al. [39] which used a transparent cell. Studies of the effect of self-breathing cathode configurations on water management are scarcely found. Mennola et al. [36] studied the effect of an open cathode design on water balance by measuring the water amount in the anode outlet. Modroukas et al. [14] observed liquid water emerging from their silicon based micro machined gas diffusion layer through stereoscopic imaging. Ous and Arcoumanis [19] studied the formation of water droplets at the GDL-channel interface in a transparent self-breathing cell. Up to date, there are no further reported studies visualizing the effect of such cathode configurations. Improvements in the spatial resolution of neutron imaging realized in the last years allowed meeting the high resolution necessary for in-plane imaging (neutron beam parallel to the membrane) [34]. This made the visualization of the liquid water distribution across the different layers of the cell possible. Furthermore, a new multi-cell setup was recently developed at the Paul Scherrer Institut [40], which allows the simultaneous characterization of up to six different cells. In this work the impact of the opening ratio (number and size of openings) onto the performance and onto the water content in the cells is studied using neutron radiography at the ICON beam line at Paul Scherrer Institut [41].

2. Experimental

2.1. Cell designs

The general cell principle and resulting cell design for the neutron radiography measurements are illustrated in Fig. 1. The cell consists of a micro-structured gold plated anodic flow field, a commercially available catalyst coated membrane MX815 from W.L. Gore & Associates GmbH, a cathodic gas diffusion layer Sigracet 35 CC® from SGL Carbon, a micro-structured 300 μm thick gold plated aluminum cathodic current collector and compression plate. The anode flow field consisted of micro-channels with a width of 0.15 mm, separated by ribs with a width of 0.55 mm. The channels were organized in an inter-digitated configuration (channels connected alternatively to the inlet and outlet manifolds). The outlet was used for purging purposes only and was closed during the experiments described here. No GDL was used on the anode side. In contrast to the usually open cathode, a closed cell configuration was chosen to allow compatibility with the existing multi-cell setup, as well as to permit controlled parameters (humidity and gas composition) on the cathode side. In order to keep a configuration equivalent to self-breathing operation, the cathode flow field design with an active area of 1 cm^2 was modified to include an empty space. At the top of this space air or oxygen was flown through the cell to generate controlled conditions of humidity. Thus, the cathode of the cell itself at the lower boundary was not affected by the forced convection, but could be operated under self-breathing conditions. The gas diffusion layer was compressed by the current collector and compression plate to a thickness of approximately 300 μm. Six different designs for the current collectors were used. The size and amount of the openings were varied for the different collectors (Table 1), leading to different opening ratios (OR).

Table 1
Design parameters of the cathodic current collectors of the studied cells and resulting opening ratios.

Size of opening [μm]	Row count [l]	Open area [mm^2]	Opening ratio [%]
600	5	1.41	15.64
600	3	0.85	9.38
600	1	0.28	3.13
300	1	0.07	1.12
150	1	0.018	0.24

2.2. Experimental conditions

During the experiments the anode was operated in dead-end mode with dry hydrogen. Initially an experiment with a constant load current with pure oxygen as well as polarization curves with air and pure oxygen was performed at 25 °C and a relative humidity (r.h.) of 50%. Furthermore, the cell performance was studied under the same environmental conditions (25 °C, 50% r.h.) for a pulsed load with respect to the required load profile. The pulse length was 10 ms, the pulse period 100 ms. In this case, the cells were operated at a current density of 500 mA cm^{-2}. In order to compare the impact of this load profile onto the water distribution within the cell, additional measurements were performed with an equivalent constant current density of 50 mA cm^{-2}. The characterization with the pulsed load and equivalent constant load was also performed at conditions with decreased humidity (ca. 20% r.h.) and increased humidity (ca. 100% r.h.) at 40 °C.

3. Results

3.1. Operation at constant current density with pure O_2

During the experiment with the constant load with pure oxygen, all cells were operating under stable conditions with loads of 100 mA cm^{-2} and 500 mA cm^{-2}. Fig. 2 illustrates the water distribution in the different cells, after a constant current of 100 mA cm^{-2} was applied for 25 min. It can clearly be observed that water is generally located on the cathode side of the cell and no accumulation of water transported back to the anode occurs. There is a uniform water distribution for cells with comparatively closed structures. On the contrary, a clear impact of the current collector structure for the cells with comparatively open structures (9–16% opening ratios) is observed, leading to a higher saturation underneath the ribs and a lower saturation underneath the openings of the current collector. The water saturation can also be analyzed further by averaging the intensity values of the radiograms for a given region. In the present work, we measure the saturation as a fraction of the total volume and not of the free pore volume.

In Fig. 3 the load profile and the resulting water saturation is illustrated for the cells with the lowest and highest opening ratio. The water saturation increases immediately after the current is