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In-operando Neutron Radiography Studies of Polymer Electrolyte Membrane Water Electrolyzers

Michael A. Hoeh[1], Tobias Arlt[2], Nikolay Kardjilov[2], Ingo Manke[2], John Banhart[2,3], David L. Fritz[1], Jannik Ehlert[1], Wiebke Lüke[1], and Werner Lehnert[1,4]

1) Forschungszentrum Jülich GmbH, Institute of Energy and Climate Research, IEK-3: Electrochemical Process Engineering, 52425 Jülich, Germany
2) Helmholtz-Zentrum Berlin GmbH, Institute of Applied Materials, Hahn-Meitner-Platz 1, 14109 Berlin, Germany
3) Technical University Berlin, Hardenberg Str. 36, 10623 Berlin, Germany
4) RWTH Aachen University, Aachen, Germany

Polymer electrolyte membrane water electrolysis cells were studied using in-operando neutron radiography, revealing insights into the gas-water distribution inside the cells. Cells were operated at current densities up to 2 A/cm² and for water flow rates ranging from 0.5 ml/(min cm²) up to 5 ml/(min cm²). The averaged gas amount in the flow channels was quantified, revealing that the ratio of gas to water inside the channels decreases for an increasing water flow rate on the anode side. This examination also demonstrates that neutron radiography is very suitable to study gas/water two-phase flow phenomena in running electrolysis cells.

Introduction

Polymer Electrolyte Membrane Water Electrolysis (PEL) is increasingly considered as an option for storing renewable energy from intermittent sources like wind or solar (1). Moreover, water electrolysis is seen as one of the few pathways available towards an electricity supply based on 100% renewables (2). PEL systems are currently scaled to larger cell areas in order to obtain megawatt systems. One of the challenges that comes along with this scale-up goes along concerns the media distribution on large cell areas. Data on the saturations of oxygen inside the porous transport layer (PTL) and the flow channels at different operating points and at different locations of a large cell area is therefore especially interesting.

In PEL, water is typically supplied via flow channels and distributed across the catalyst layer through the PTL. At the catalyst layer, gas evolves which is transported back through the PTL and discharged into the flow channels. From the flow channels, it is transported out of the cell as a two-phase mixture with the feed water. This evolution of oxygen and hydrogen in the surrounding water is investigated in this study in-operando inside operating PEL cells using neutron radiography. In the past, radiography methods were applied to visualize water management issues (3) and liquid water formation in polymer electrolyte membrane fuel cells (PEFC) (4), as well as the evolution of carbon dioxide in direct methanol fuel cells (DMFC) (5). Radiography has recently been applied to PEL, studying the evolution of water thicknesses in a PEL cell over time (6) and the discharge of bubbles into the flow channel (7).

In this work, operating PEL cells are examined using neutrons at BER II (Helmholtz-Zentrum Berlin). The neutron radiography setup used for the experiment allowed inspecting areas of several cm² with an optical resolution of about 50 µm. The ratio of water to gas was quantified for different operating conditions.

Experimental Method

The PEL cells used the neutron measurements exhibited an active area of 17.6 cm^2. All cells utilized membrane electrode assemblies (MEAs) procured from Solvicore, which consisted of a Nafion N117 membrane (DuPont) coated with a cathode catalyst layer consisting of platinum and an anode catalyst layer containing iridium oxide. On the anode side, PTLs composed of sintered titanium powder were used, while Toray carbon paper (TGP-H-120) was used on the cathode side of all cells. Graphite separator plates with a machined flow field structure were used with a meander-shaped single channel on the anode and equally-spaced parallel channels on the cathode, as shown in Fig. 1. The channels were 1.5 mm wide and 1.5 mm deep. The PEL cells were operated at ambient pressure and 80°C.

The measurements were carried out at the CONRAD-2 (8, 9, 10) beamline at the BER II neutron source at Helmholtz-Zentrum Berlin (HZB). The experimental setup comprised a detector system (sCMOS camera "Andor NEO") with a pixel size of 22 µm and a scintillator screen (6LiFZnS) with a thickness of 100 µm. A beam collimation ratio (L/D) of 350 was used. The PEL cell was placed in the neutron beam with the MEA being perpendicularly oriented to the beam. Images were continuously acquired, with exposure times of one and five seconds.

The neutron beam is directed to the cell, and is attenuated inside the cell in dependence on the materials employed in cell construction. Neutron radiation is very sensitive for light elements like hydrogen. This means that most of the neutrons were attenuated by the water that is supplied to the cell. In order to obtain a measure for the change in the amount of gas between two settings, all images were divided by a reference image. This reference image was obtained from a cell that was supplied with water, but without electrolysis operation, so without gas in the cells originating from electrolysis. This way the reference image represents the condition that the flow channels are filled with water only, and no gas portion. The Lambert-Beer law was used to calculate the changes in the gas amount in the cells. An attenuation coefficient of 5.2 cm^{-1} was determined in previous calibration measurements for a defined distance of 5 cm between the cell and the detector. In case there is more water present in the cell compared to the reference image, these areas will appear darker in the image. Areas with less water will appear in light gray values, respectively.

Cell Setup

The cells were operated with water flow rates of 0.5, 2, 3.5 and 5 ml/(min cm^2). The water was supplied to the bottom of the cell, while the outlet of the flow channel was situated at the top of the cell (cf. Fig. 2). Only the anode side was supplied with water, the cathode was not supplied with water. The current densities were chosen as 0.1, 0.4, 0.8, 1, 2 A/cm².

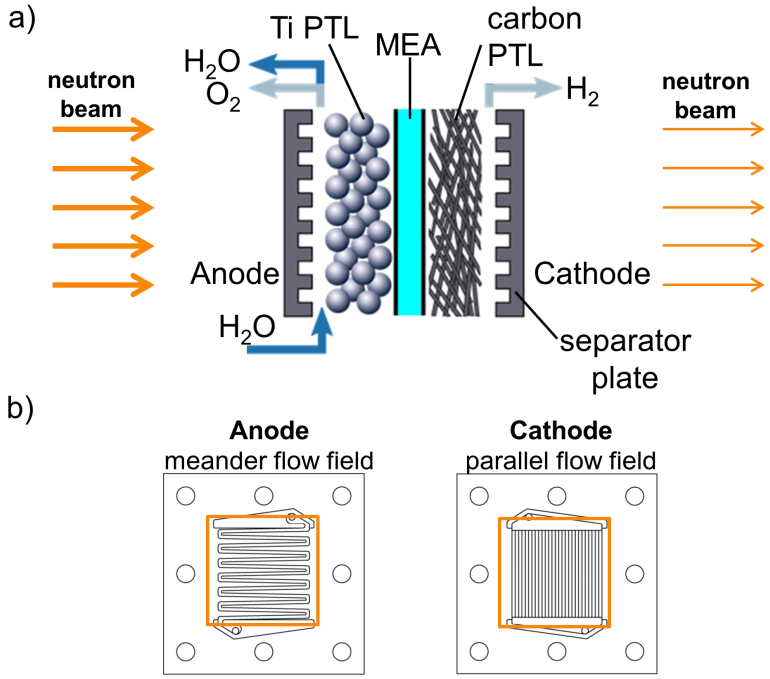

Figure 1. Sketch of the PEL cell showing the components separator plates, titanium PTL, MEA, carbon PTL. The neutron beam is directed at the cell in through-plane direction, and the neutron beam transmitted through this structure is detected behind the cell
(b) Flow field structures used for the measurements, with a meander-shaped flow field on the anode side and a parallel channel flow field on the cathode side. The yellow boxes indicate the areas visualized in the experiment.

Results

Fig. 2 shows an image series of neutron radiographs taken at different current densities at a flow rate of 0.5 ml/(min cm²). At the current density of 0.01 A/cm², the structural characteristics of the anode channel can be observed, but the gray values of the channel area only slightly differs from the one in the other parts of the cell, indicating that primarily water is transported in the flow channel. In Fig. 2b) at a current density of 0.1 A/cm², the produced gas can already be observed in the flow channel, with a rising portion of the gas to the top cell outlet. This portion of the gas increases for increasing current densities. At 1 A/cm² primarily gas is transported in the channel, already at the water inlet of the cell. For Fig. 2c) and d) dark horizontal lines are observable, which represent water accumulating in the parallel channels of the cathode flow field. The cathode was not supplied with water from outside, so the water accumulating in the cathode flow channels originates from the water permeated through the membrane from the anode side due to electro-osmotic drag.

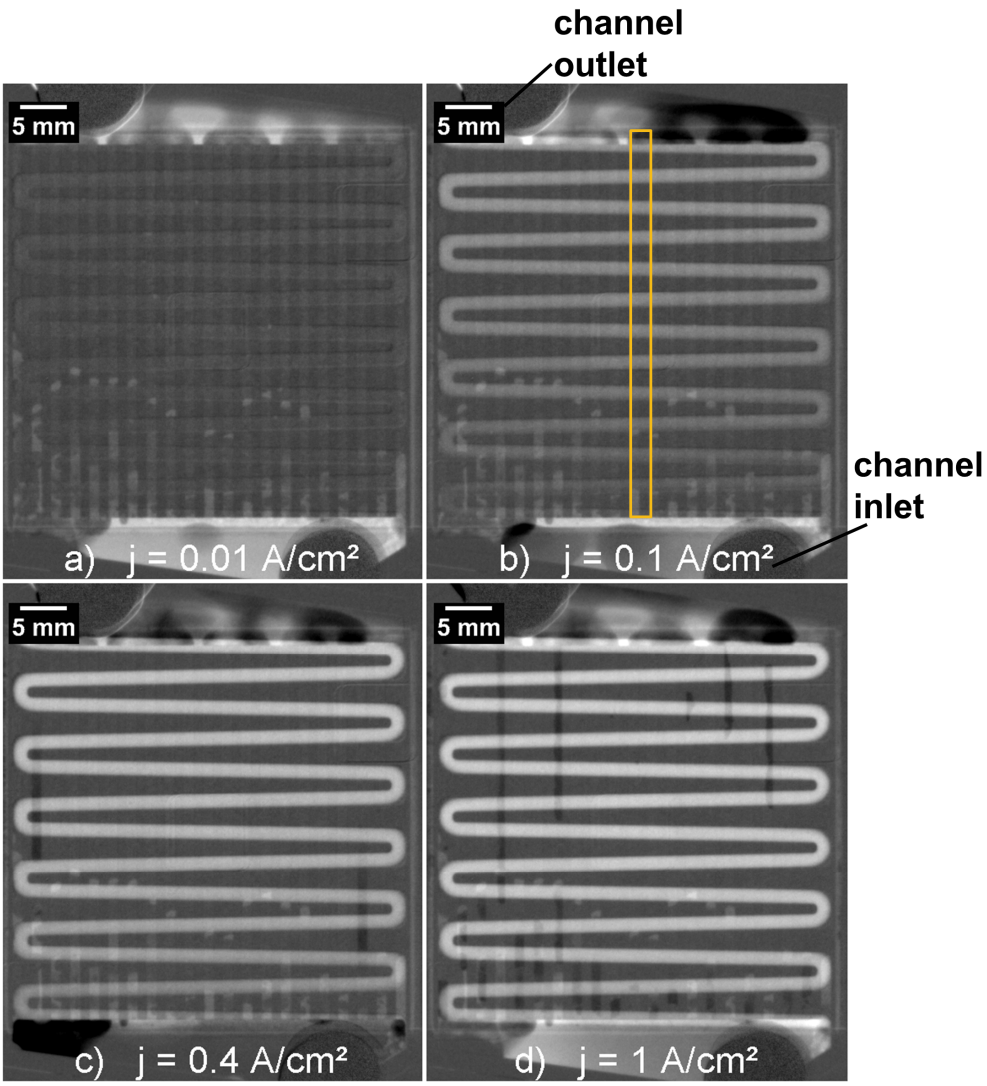

Figure 2. Image series of neutron radiographs of the cell at current densities of a) 0.01 A/cm² b) 0.1 A/cm² c) 0.4 A/cm² d) 1 A/cm² and anode water flow rate of 0.5 ml/(min cm²) from the channel inlet (bottom of the cell) to the channel outlet (top of the cell). The box indicated in yellow in b) represents the area considered for the plot in Fig. 3

These observations from the image series in Fig. 2 were further quantified using the Lambert-Beer's law in order to determine the gas quantities within the flow channel. This was accomplished by selecting a region on the radiographs of the cell, horizontally from the cell center and vertically ranging from the bottom of the cell to the top of the cell (yellow box indicated in Fig. 2b), and averaging laterally in horizontal direction. This allowed examining the change in the gas amount in the channel from a position close to the channel inlet to a position close to the channel outlet. The results of this examination are shown in Fig. 3 at current densities ranging from 0.1 A/cm² to 2 A/cm² for water flow rates of a) 0.5 ml/(min cm²), b) 2 ml/(min cm²), c) 3.5 ml/(min cm²), d) 5 ml/(min cm²). The gas ratio expresses the share of the gas in the water-filled channel, so a ratio of 0.8 describes the fact that 80% of the channel is filled with gas, 20% with water. It can be seen that for an increase in water flow rates through the flow channel from a) to d), the amount of gas in the water/gas two-phase flow decreases.

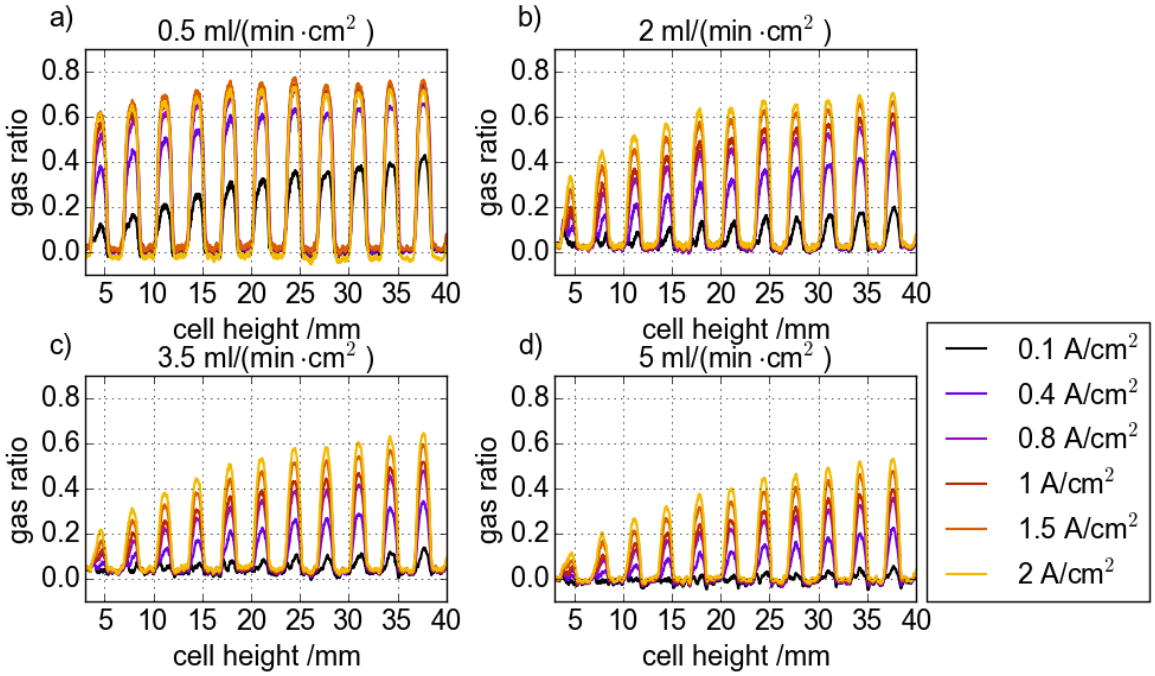

Figure 3. Gas ratio in the channels and ribs of the flow field at current densities ranging from 0.1 A/cm² to 2 A/cm² for water flow rates in the channel of a) 0.5 ml/(min cm²), b) 2 ml/(min cm²), c) 3.5 ml/(min cm²), d) 5 ml/(min cm²). The left of the plot marks the channel inlet (zero cell height), while the right of the plot represents the area close to the channel outlet. The box in Fig. 2b) indicates the region used to obtain this data.

Conclusion

This study employed neutron radiography in order to study the gas amount in running Polymer Electrolyte Water Electrolysis cells, operating at current densities up to 2 A/cm² and for water flow rates ranging from 0.5 ml/(min cm²) up to 5 ml/(min cm²). The gas amount in the flow channels was quantified, revealing that the ratio of the produced gas to water inside the channels decreases for an increasing water flow rate on the anode side. This examination also highlights that neutron radiography represent a valuable tool that allows for analyzing the gas-water distribution inside running PEL cells, and also allows quantifying the gas-water proportions. The results obtained provide information for the modeling of two-phase flow in PEL cells, and further reveal relevant insights for designing cell components with facilitated gas removal.

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