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Short Communication

Water management in novel direct membrane deposition fuel cells under low humidification



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ABSTRACT

Polymer electrolyte membrane fuel cells (PEMFCs) fabricated by direct membrane deposition (DMD) were shown to work even at dry conditions without significant deterioration of the membrane resistance. Here, in situ neutron radiography is used to investigate the water management in those fuel cells to uncover the phenomena that lead to the robust operation under low humidification. A constant level of humidification within the membrane electrode assembly (MEA) of a DMD fuel cell is observed even under dry anode operation and 15% relative humidity on the cathode side. This proves a pronounced back diffusion of generated water from the cathode side to the anode side through the thin deposited membrane layer. Over the entire range of polarization curves a very high similarity of the water evolution in anode and cathode flow fields is found in spite of different humidification levels. It is shown that the power density of directly deposited membranes in contrast to a 50 μm thick N-112 membrane is only marginally affected by dry operation conditions. Water profiles in through-plane direction of the MEA reveal that the water content in the DMD fuel cell remains steady even at high current densities. This is in contrast to the N-112 reference fuel cell which shows a strong increase in membrane resistance and a reduced MEA water content with raising current densities. Thus this new MEA fabrication technique has a promising perspective, since dry operation conditions are highly requested in order to reduce fuel cell system costs.

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Introduction

Direct membrane deposition (DMD) is a novel MEA fabrication method, where the membrane layer of a PEMFC is directly deposited onto its anode and cathode gas diffusion electrodes [1]. This MEA fabrication method enables very low membrane resistances below $13 \text{ m}\Omega \text{ cm}^2$ and therefore high fuel cell power densities beyond 4.0 W/cm^2 with H_2/O_2 as feed gases at $300 \text{ kPa}_{\text{abs}}$ and beyond 1.3 W/cm^2 under stoichiometric 1.2/2.0 H_2/air operation at $300 \text{ kPa}_{\text{abs}}$. The high performance was linked to a very low membrane thickness of about $12 \mu\text{m}$ and an improved interface between catalyst layer and the membrane itself [1,2]. Despite the high power densities another significant difference was identified compared to conventionally cast membranes: The cell polarization measurements in the work of Klingele et al. [1] reveal that the ionic resistance increased only slightly at low humidification. Even when the humidification of the reactant gases was reduced to zero, the maximum cell power dropped only by about 10%. This finding diverged from the known characteristics of fuel cells with conventional Nafion® membranes [3–5] and is therefore of high interest for further investigation. Even though dry operation of fuel cells with Nafion® membranes was reported in literature before, a more significant drop in cell power density between 20 and 40% was usually observed [3]. To enable dry fuel cell operation by self-humidification, complex composite structures such as thin Nafion/Pt/SiO₂ [6], Nafion/Pt-clay [7] or sulfonated poly(ether ether ketone) (SPEEK)/Pt-Zirconia [8] composite membranes have been proposed. Klingele et al. demonstrated the operation of a directly deposited pure Nafion membrane without any external humidification. This finding was attributed to a strongly facilitated back diffusion of water from the cathode to the anode side through the about $12 \mu\text{m}$ thin directly deposited membrane. This back diffusion was believed to humidify the membrane sufficiently even under dry conditions and at high current densities. To prove this hypothesis, this work presents in situ neutron imaging of fuel cell operation with dry anode feed gas to analyse the amount of water in the MEA under low humidification.

Neutron imaging is well suited to investigate water evolution and has been widely employed in in situ fuel cell experiments [9–14]. The strong attenuation of neutrons in water enables imaging liquid water evolution within a fuel cell in operation. The neutron-based analysis of various fuel cell operation conditions [15], flow field designs [16] or gas diffusion materials [17] was reported in literature previously using conventionally cast membranes. Water back diffusion, even in a thick Nafion® N-117 membrane, was recently investigated by Iranzo et al. [18]. These studies motivate the use of neutron imaging for the characterization of the water distribution in DMD fuel cells.

In situ neutron radiography

In situ neutron radiography experiments were performed at the neutron beamline “ANTARES” at Heinz-Maier-Leibnitz Zentrum (MLZ) at the Technische Universität München

(TUM). The setup was described in detail by Calzada et al. [19] and is shown schematically in Fig. 1a). The neutron beam coming through a pinhole from the reactor can be used for imaging purposes in the two separate beam chambers. The fuel cell, shown in Fig. 1b), was located in the first measurement chamber (blue area in Fig. 1a). We used a specifically designed aluminium fuel cell fixture (see Fig. 1b). This ensured sufficient transparency of the PEMFC for the beam. The fuel cell was operated from outside the beam chamber via 5 m long heated gas lines. This ensured that the sensitive electronics of the fuel cell tester could be placed outside the beam chamber and thus were not damaged during the experiments. For the neutron experiments, the fuel cell was placed in-plane with the beam direction to observe the dynamic water evolution inside its anode and cathode flow channels separately (Fig. 1b). A 18 mm pinhole was used, resulting in an L/D ratio of 500 at the sample position. The effective pixel size was about $27 \mu\text{m}$ and was determined experimentally from the known geometry of the micro-milled gas flow channels of $500 \mu\text{m}$ width. For the neutron images during polarization measurements shown in Fig. 3, an integration time of 60 s was chosen to enhance water sensitivity. To track the water evolution as dynamically as possible during a steady potential, an integration time of 5 s was chosen. Due to this short integration time, the recorded radiographies at steady state conditions were rebinned to a resolution of $137 \mu\text{m}$ in order to achieve a sufficient signal-to-noise ratio as described elsewhere [20].

To extract the actual water generation in the flow channels, the neutron absorption of the dry fuel cell was taken as reference. The water profiles shown in Figs. 2 and 3 were generated by summarizing the water signal in the fuel cell. The offset of the CCD detector (Andor, Ikon L, 2048×2048 pixels) was eliminated by subtraction of a dark image with closed beam shutter. Gamma spots were corrected in the post-processing. Before each in situ experiment a neutron image under OCV conditions was recorded to exclude a falsification by residual water from previous operation.

Sample preparation and fuel cell operation

To fabricate the DMD fuel cell, one layer of Nafion® dispersion (1 part Nafion® D2020 mixed with two parts 1-propanol) was inkjet-printed onto 5 cm^2 anode and cathode gas diffusion electrodes (GDEs) purchased from Paxitech SAS. The GDEs were loaded with $0.5 \text{ mg}_{\text{Pt}}/\text{cm}^2$ at 70% Pt/C and contained Nafion® as ionomer. The thickness of each catalyst layer was about $10 \mu\text{m}$ (measured by scanning microscope imaging on MEA cross-sections), the thickness of each gas diffusion layer (in the compressed state) was approximately $200 \mu\text{m}$, according to the manufacturer. The fuel cell was assembled by placing the two GDEs covered by the printed membrane layers face to face. A $50 \mu\text{m}$ thin PTFE foil was used as subgasket to prevent parasitic currents and gas crossover occurring at the edges of the GDEs. The subgasket reduced the active fuel cell area to 4 cm^2 . The fuel cell (shown in Fig. 1b) was tightened with a torque of 1 Nm. A detailed description of the sample preparation including the printing process was published previously [1,2]. As reference, we used an N-112 Nafion® membrane, coated with catalyst layers of identical Pt loading

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