Identification of liquid water constraints in micro polymer electrolyte fuel cells without gas diffusion layers

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\textbf{A R T I C L E  I N F O}

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\textbf{A B S T R A C T}

A simplified, miniaturized polymer electrolyte fuel cell without gas diffusion layers was investigated under operation by neutron radiography. By visualizing liquid water, it was possible to identify limiting effects, which are directly related to the simplified construction principle. Depending on the operation conditions, undesired water accumulation either in particular micro-channels or on the cathode catalyst layer as well as drying of the anode catalyst layer was observed. As a consequence, the design of a fuel cell without gas diffusion layers must take into account these limitations visualized by neutron radiography.

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1. Introduction

Polymer electrolyte fuel cells (PEFCs) are suggested to power devices of different scales, ranging from pacemakers to vehicles. One advantage of PEFCs is the high efficiency due to direct conversion of chemical energy, stored in hydrogen, to electrical energy. In comparison to batteries, high energy densities are predicted for fuel cell systems including hydrogen storage\cite{1}, a fact which is of interest for portable electronic devices.

Simple down-scaling of fuel cell assemblies, originally developed for automotive application, is one way to obtain a miniaturized fuel cell\cite{2–5}. Precise assembly and negative scaling effects create challenges that could hamper industrial production. One solution might be membrane-based designs, where micro-patterned parts are fixed onto the membrane\cite{6–8}. Taking into account the proposed production processes, where the fuel cell stack is assembled in foil-type planar shape, designs without gas diffusion layer\cite{9,10} become even more interesting due to easier assembly, easier tightening, and reduced number of parts.

Only few of the presented micro polymer electrolyte fuel cell concepts are able to deliver current densities at which mass transport limitations in technical fuel cells can occur\cite{11}. However, the influence of liquid water is already crucial at lower current densities. Beneficial effects are remarkable, such as improved proton conductivity in the membrane\cite{12} as well as the formation of triple-phase boundaries in the catalyst layer, which are needed for the hydrogen oxidation and the oxygen reduction reactions, respectively. Due to water being removed from the anode by the electrochemical drag\cite{13}, gases have to be humidified prior to being fed into the fuel cell.

Contradicting this statement, Nguyen and Chan\cite{11} point out that space constraints in micro fuel cell systems end up in designs without an active water management; i.e. developers forego an external humidification and expect the anode to be humidified by diffusion of product water from the cathode. Metz et al.\cite{14} realized a passive water management for a micro fuel cell by using capillary micro-structures. They only report about positive effects on the cathode side when the operation conditions are changed and drying can be avoided.

However, liquid water can reduce the access of hydrogen and oxidant to the catalyst sites, especially when the fuel cell is not optimized for liquid water removal. Although the Peclet number, which is typically low for miniaturized PEFCs, may be an indication for insufficiencies in water removal\cite{11}. This effect might be detrimental in self-breathing concepts, as for example suggested by Xia and Chan\cite{15} or Modroukas et al.\cite{16}, which rely on free convection for oxygen supply and water removal on the cathode side. Modeling predicts an increased power density for decreasing channel dimensions down to 5 \(\mu\)m because the effective area of the...
electrode is increased for comparable membrane size and reactant flows [2,17]. Experimental validation of such results is challenging due to pressure drop and water accumulation [1]. Otherwise, Cha et al. [18] could show that, in the case of a miniaturized fuel cell including a gas diffusion layer, small channels can have a positive influence on the performance.

Up to now many concepts for micro PEFC have been published, but a profound understanding about limiting effects under operation has to be further developed. The method of choice to visualize liquid water in a PEFC under operation is neutron radiography [19]. The concentration of gaseous water is too low to be visible in neutron radiograms. Based on the improved resolution of a new detector system [20], even in-plane radiography can be carried out, i.e. in parallel direction to the membrane plane [21]. Hence we can fulfill the demand of Nguyen and Chan [11] and apply a method to visualize liquid water in a miniaturized polymer electrolyte fuel cell while operating. The employed miniaturized PEFC was developed at Paul Scherrer Institut (PSI) and it proved its ability to deliver power densities in a similar order of magnitude as reported for technical polymer electrolyte cells with several square centimeters of active area [22]. The cell is operated without gas diffusion layers (GDL). The advantages of this concept are easier assembly, smaller number of parts and easier sealing due to no porous parts being employed in the fuel cell. However, the functions of a GDL, such as fine gas distribution and electrical connection of the active sites, have to be fulfilled by the micro-channels and the catalyst layer.

2. Experimental

The micro PEFC was assembled by sandwiching a catalyst-coated membrane (CCM) between two micro flow fields. CCMs including three different membranes were employed: Nafion 112, Nafion 211, and Gore Primea 5710. Each CCM contained platinum loadings of 0.4 mg/cm² (cathode) and 0.1–0.15 mg/cm² (anode).

The micro-patterning of glassy carbon has been published elsewhere [23–26]. The three-step process offers a great flexibility to produce various flow field geometries. For these experiments, a meander consisting of seven parallel channels (type A) was applied as well as a meander with merging channels (type B), whose number is reduced along the flow field from eleven to three channels.

The trapezoid-shaped channels were 110 μm deep and 100 (top) to 60 μm (bottom) wide, with a channel-to-rib-ratio of 5/3. The flow fields were surrounded by gold-coated aluminum end plates, in which the functions of electrical contact, gas supply to the microstructures, temperature control, and mechanical compaction are combined. The pressure in contact areas was adjusted by four bolts to a value of 2.2 MPa.

The fuel cell, situated in the neutron beam, was operated by a home-build fuel cell test bench, which permanently controls and monitors gas flows, gas humidification, pressure, temperature, current, and voltage. AC impedance at 1 kHz of the fuel cell was monitored by a Tsuruga 3566 measurement device in order to identify increases of the high frequency resistance caused by drying effects in the membrane.

The neutron radiography experiments were carried out at the ICON [27] beamline at the neutron spallation source SINQ [28] of PSI. For our investigations, through-plane as well as in-plane neutron radiography were utilized, i.e. perpendicular and parallel to the membrane plane (Fig. 1).

Different imaging setups were used for in-plane and through-plane imaging. For in-plane imaging, a setup with anisotropic resolution [21] was used, due to different resolution requirements according to the direction (the highest resolution is required in the direction across the membrane). The detector pixel sizes as well as the effective spatial resolution of both setups are listed in Table 1. The effective resolution is defined as the Full Width at Half Maximum (FWHM) of the Line Spread Function (LSF) of the imaging setup.

Radiograms were referenced with a radiogram recorded at open circuit voltage (OCV) under dry conditions [19]. Additionally, selected results were referenced with radiograms recorded under D₂/D₂O operation of the fuel cell to exclude misinterpretation possibly caused by membrane swelling. In a referenced in-plane radiogram, the water content was obtained by quantitative evaluation of its gray scale profile.

Recording SEM micrographs and cross section profile analysis of the tested catalyst-coated membranes was carried out using a Zeiss Gemini 55 scanning electron microscope.

3. Results

3.1. Water accumulation in channels

Our micro PEFC shows unsteady behavior when flow fields in meander shape with parallel channels are employed. Irregular

<table>
<thead>
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<th>Table 1</th>
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<tr>
<td>Detector pixel size and effective spatial resolution of the applied neutron radiography setups.</td>
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<tr>
<td></td>
</tr>
<tr>
<td>Pixel size/μm</td>
</tr>
<tr>
<td>Through-plane</td>
</tr>
<tr>
<td>In-plane</td>
</tr>
</tbody>
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Fig. 1. Measurement principle for through-plane and in-plane neutron radiography on a GDL-less micro PEFC. For in-plane neutron radiography, the rectangle indicates the area that is analyzed to obtain a water profile.
water removal from the cathode side, pressure drop fluctuations on the cathode side, and cell voltage fluctuations are observed [22]. Through-plane neutron radiography is the method of choice to investigate these characteristics that we assumed to be caused by water accumulation. We recorded a series of neutron radiograms in order to assess localized water accumulation within the flow fields or particular channels (Fig. 2). The time difference between recording radiograms A1 and A2 is 12 min. One can easily see that two particular cathode channels are filled with liquid water in A1, while after 12 min one of these channels is drained. In the meantime, a decrease in pressure drop and an increase of cell voltage took place. Quantification of the water signal reveals that the cathode channels, which are 110 μm deep, are completely filled with water and, therefore, bypassed by the cathode gas flow. We assume that mass transport is hindered in the flooded channels, while the reaction continues in non-flooded areas of the cell, until water accumulates there too, leading to a draining of the completely filled channels due to the increased pressure drop. Chen et al. [29] report about the formation of a plug/bubble flow regime in micro-channels. Our observations of batch-wise water removal from the fuel cell support their evidence. The two radiograms B1 and B2 in Fig. 2 were recorded in a similar series, but with merging micro flow fields. Irregular water removal as well as pressure drop and cell voltage fluctuations were not observed in this case. In contrast to series A, no water accumulation in particular channels is visible, but a rather smooth distribution of water over almost all channels. The bypassing effect taking place in the parallel meander does not take place here due to the reduced total cross section of the channels towards the outlet, and the gas velocity remains therefore similar as at the beginning of the channels.

As a consequence, one has to keep in mind when building a micro fuel cell with active water removal that the right balance between pressure drop and flooding has to be found. In our case, a further reduction of channel number would significantly increase the pressure drop [22] while increasing the number of channels increases the bypassing effect as described here. The merging flow field turns out to combine both, moderate pressure drop (e.g. 450 mbar for a dry 20 sccm O2 flow) and a steady liquid water removal from the cathode.

These results have to be discussed in the context of modeling results [2,17,18], which predict improved fuel cell performance if the channel dimensions are reduced down to widths of 5 μm and below. Decreasing the channel size causes an increased number of channels per area. In order to keep pressure drop losses small, a high number of parallel channels is needed in such flow field concepts. Too many parallel channels may result in unsteady water removal and, hence, unsteady fuel cell performance. A compromise has to be found between these two negative effects. Alternatively, intelligent flow field geometries must be developed in order to apply micro-channels in the 5 μm range.

3.2. Water accumulation on the cathode catalyst layer

Recording current–voltage curves on our micro PEFC results in an abrupt voltage decrease at a reproducible limiting current density. For a CCM based on a Nafion 112 membrane, this limiting current density can be observed at an approximate value of 1.05 A cm$^{-2}$ [22]. For different CCMs, a similar behavior could be observed, but at different current densities. For a CCM containing Nafion 211 as membrane, the limiting current density reaches a value of 1.45 A cm$^{-2}$, and 1.8 A cm$^{-2}$ for a Gore Primea 5710, respectively.

In-plane radiograms (Fig. 3) recorded at A cm$^{-2}$, employing the three different CCMs, reveal that the water layer is only formed at the surface of the catalyst-coated Nafion 112 membrane in the micro-channel (Fig. 3A). For the other two CCMs neither in the radiogram itself nor in the water profile, a water layer is visible at the same current density (Fig. 3B and C). The water layer appears
only when the current density is increased towards the typical limiting current density. The formation of a water layer on the catalyst induces oxygen mass transport limitation to the active sites on the cathode. As soon as the limiting current density is reached, the mass transport governs the behavior of the fuel cell resulting in an abrupt decrease of cell voltage.

We assume that the reason for the differences in limiting current density is correlated to the different creeping behavior of the respective membrane. Creeping takes place as a consequence of membrane swelling when hydrated, and of compression due to clamping. In our fuel cell assembly without gas diffusion layers, there is no soft material which compensates the clamping pressure applied to the membrane. Thus, the membrane creeps into the only space available, i.e. the micro-channels. The CCM including a Nafion 112 membrane, which is the thickest of the three employed membranes, shows the most pronounced creeping, followed by the CCM with the Nafion 211 membrane, and finally the Gore CCM. This is depicted in the SEM micrographs in Fig. 4 and the data listed in Table 2.

The more creeping is taking place, the more mechanical stress affects the catalyst layer, resulting in more rupturing and larger area defects (Fig. 4). The pristine catalyst layer is hydrophobic. For all three CCMs, we recorded a contact angle (water/air) in the range of 145–147°, which is in agreement with published data [30]. When the catalyst layer is ruptured, more hydrophilic membrane material is exposed towards the gas channel. This hydrophilic surface attracts water and at high current densities a water layer is formed. Results obtained by Yu et al. [30] suggest a similar effect. Using environmental scanning electrode microscopy (ESEM), they investigated a membrane electrode assembly (MEA) after operation in a technical PEFC. Contact angles measured as low as 33° indicate that ionomer material is present at the outer surface of the catalyst layer.

### Table 2

<table>
<thead>
<tr>
<th>CCM</th>
<th>Original thickness/μm</th>
<th>Maximum thickness under channels/μm</th>
<th>Thickness under ribs/μm</th>
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<tbody>
<tr>
<td>Nafion 112</td>
<td>50</td>
<td>79</td>
<td>16</td>
</tr>
<tr>
<td>Nafion 211</td>
<td>25</td>
<td>32</td>
<td>14</td>
</tr>
<tr>
<td>Gore Primea 5710</td>
<td>18</td>
<td>20</td>
<td>15</td>
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</table>

### Fig. 4

In-plane water profiles of a micro fuel cell including type B flow fields and containing a catalyst-coated Nafion 112 membrane at different operation points (cell temperature $T_{\text{cell}}$, cell voltage $E_{\text{cell}}$, $\lambda_{\text{H}_2} = 1.7$, $\lambda_{\text{O}_2} = 2.1$, $j = 1 \text{ A cm}^{-2}$. 

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Fig. 3. Comparison of radiograms of fuel cells including type B flow fields and operating with different CCMs: Nafion 112 (A), Nafion 211 (B) and Gore 5710 (C) at $T_{\text{cell}} = 70^\circ \text{C}$, R.H. = 80%, $\lambda_{\text{H}_2} = 1.7$, $\lambda_{\text{O}_2} = 2.1$, and $j = 1 \text{ A cm}^{-2}$. The gray scale profiles on the right hand side indicate a more pronounced water layer next to the CCM for fuel cell A.
3.3. Temperature and gas humidification effects on cell performance

Parameter screening showed that the optimum operation point is at a cell temperature of 50 °C and a relative humidity (R.H.) of 80% for the gases when utilizing a Nafion 112 CCM in the micro fuel cell. Gas humidification does not have a substantial influence at this relatively low temperature, because most of the water present in the fuel cell originates from the reaction and not from the external humidification of the gases. Obviously, this observation is also expressed by the water profiles recorded at a cell temperature of 50 °C (Fig. 5A and B). No significant difference in terms of water distribution is visible for relative humidities of 35% and 80% R.H. The same holds for the cell voltage, which amounts to 587 mV at a current density of 0.6 A cm⁻² for both cases, and the high frequency resistance that remains constant at 152 mΩ cm². Only when the anode humidification is reduced to 0% relative humidity, significant losses are observed in combination with a drastically increased high frequency resistance.

Increasing the temperature to 70 °C at 80% R.H. results in a reduced cell voltage (558 mV) and in reduced amounts of liquid water in the cathode and the anode channels (Fig. 5C), while the high frequency resistance does not increase significantly. The higher water saturation pressure at higher temperatures causes a drying effect on the anode side. In contrast to a PEFC with gas diffusion layers, the catalyst area is directly exposed to the gas flows and, consequently, more susceptible towards drying effects. A reduction of the gas humidification to 35% (Fig. 5D) results in an even lower cell voltage (532 mV) and in an even lower water content on the anode side and in the bulk membrane. Additionally, we could observe an increase of the high frequency resistance to 190 mΩ cm². The effect of too dry anode conditions can be compensated by supersaturation of the hydrogen stream to a relative humidity of 115% (Fig. 5E). The water content for the anode channels is comparable to the conditions of a cell temperature of 50 °C. Finally, our hypothesis is proven by the fact that the cell voltage has increased to 584 mV again, at a current density of 0.6 A cm⁻². The increase of the gas humidification above 35% on the cathode side does improve the performance of the cell, while a further increase of the gas humidification on the anode side above 115% results in condensation already at the gas inlets of the cell and, as a consequence, in non-stable operation of the fuel cell.

During operation of PEFCs it is sometimes observed that water accumulation even on the anode side takes place if the hydrogen stoichiometry is only slightly above 1. However, we did not encounter such effects when reducing the hydrogen stoichiometry in these experiments and were not able to avoid drying of the anode catalyst layer.

3.4. Hysteresis effects during current–voltage curve recording

During the recording of a current–voltage curve it is important to remain sufficiently long at each data point to allow the fuel cell to equilibrate. We made the experience that an equilibration time of 2 min is enough for our micro fuel cell. In order to demonstrate the impact of a too short equilibration time, current–voltage curves with varying equilibration time and neutron radiograms were recorded simultaneously. One of these series is illustrated in Fig. 6 with an equilibration time of 30 s for each current density value at 70 °C and a relative humidity of 50% for anode and cathode gas. The resulting hysteresis effects can be easily observed in the current–voltage curve. At the limiting current density of 1.4 A cm⁻², the water layer on the cathode catalyst, as discussed in Section 3.2, can be observed, as well as liquid water on the anode side originating from water diffusion through the membrane. After decreasing the current density again, excess water on the cathode side is still present and causes mass transport limitations. The latter results in a decreased value for the cell voltage at a current density of 1 A cm⁻² (F), compared to the previous measurement, when the current density was increased to 1 A cm⁻² (D).
the current density is further decreased in one step to 0.5 A cm⁻² (G), even more liquid water is present on the anode side than it was detected while recording data point C. Now, this effect prevents the anode from temporary drying, and a higher cell voltage is obtained. Under steady-state operation, utilizing the CCM with a Nafion 211 membrane we could achieve a cell voltage of 497 mV at 1 A cm⁻² resulting in a power density of 497 mW/cm². Hence, our results show that false power density values are obtained when current–voltage curves are recorded without sufficient equilibration time. Additionally, these results reveal the importance of liquid water formation, inducing a mass transport limitation on the cathode side and drying effects on the anode side of the miniaturized and simplified PEFC.

3.5. Comparison of referencing methods for neutron radiography on fuel cells

As a standard procedure, a radiogram recorded under certain humidity conditions is referenced to a radiogram recorded under ‘dry’ conditions and at open circuit voltage (OCV)[19]. As a consequence, the membrane may appear thinner as in radiograms recorded under humid conditions because then the membrane material swells due to water uptake. Hussey et al. [31] could observe this effect when recording in-plane neutron radiograms of a fuel cell employing a Nafion 117 membrane (dry thickness 178 μm). We were able to exclude such misleading effects the following way: A reference radiogram was recorded at the very same operation conditions as the radiograms to be referenced, but in this case the fuel cell was operated with deuterium (D₂) as feed gas, and both gas flows were humidified with heavy water (D₂O). No significant difference could be observed in terms of fuel cell performance. Thus, we assume a similar swelling behavior of the membrane as under H₂/H₂O operation. The cross section of cold neutrons with D₂O is one order of magnitude smaller than the one with H₂O [32]. This fact offers the possibility that operation of the cell with D₂ as reactant and D₂O as humidifying agent can be used as reference state. Comparable water profiles show that both radiograms are close to being identical (Fig. 7) despite being referenced with different methods. Thus, we can exclude swelling effects in our in-plane neutron radiography experiments.

4. Discussion

PEFCs are usually designed to be operated with air. Oxygen was used instead of air to avoid another source of mass transport limitation possibly caused by diffusion of oxygen in air. Further effects could make the interpretation of our results difficult, e.g., the pressure drop within the flow fields would increase by a factor of five. The authors are aware that the gained results may not be transferred directly to polymer electrolyte fuel cells operated with air. However, selected fuel cell experiments with air using merging flow fields and a Gore CCM showed comparable effects, such as the limiting current density or the low optimum operation temperature.

The CCM protrusion into the channels as described in Section 3.2 may influence the results gained from neutron radiography, too. An increase in pressure drop is taking place by decreasing the cross sectional area of the micro-channels, but a complete channel blockage can be excluded regarding the fact that even the CCM containing a Nafion 112 membrane protrudes into the channel only to about 30% of the channel depth. Therefore, the observed filling and draining effect in Section 3.1 is a water management issue because the reduced volume of the channels supports faster water filling of particular channels. If the applied micro–channels were less deep, one must consider this effect. For in-plane radiograms, the protruding CCM shifts the interface, where water accumulation takes place as described in Sections 3.2, 3.3 and 3.4 into the channels. This effect must be considered analyzing the dimensions of the radiograms, which agree with the dimensions obtained post-mortem and given in Table 2. The water profiles in Fig. 4 reflect these dimensional effects, a slight shrinking of the membrane with low humidification can not be excluded, but it does not affect the qualitative conclusion gained from the result.

One of the limitations of neutron radiography is that we are only able to map the water distribution in the fuel cell in two dimensions. In through-plane radiography information about the amount of liquid water can only be obtained from the signal intensity, but discrimination between water on either of the two sides is not possible. Our in-plane radiograms show clearly that most of the signal originates from water on the cathode side.

In in-plane radiography, only integral quantification over all channels of the cathode or the anode side is possible. However, inhomogeneities of the water distribution are clearly visible along the channels in Fig. 2. Hence, integral water profiles, as shown in Fig. 4, can only be compared qualitatively, while the determination of the amount of water remains difficult.

We have to face the situation that the removal of the gas diffusion layers in a fuel cell results in new limitations, which are caused by the fact that other particular parts in the fuel cell have to fulfill the typical functions of a GDL. In our micro fuel cell, the micro-channels must provide the gas distribution while the catalyst layer itself has to conduct the electrons in-plane to or from the active catalyst sites. Both limitations can be minimized by an intelligent design of micro-channel and rib geometries. But other functions of the GDL, such as the compensation of the clamping pressure acting on the membrane, the removal of the water from the backside of the catalyst layer or the protection of the anode catalyst layer from complete drying, can not be achieved in this micro fuel cell concept.

The influence of temperature and gas humidification, respectively, demand an active water management, especially when the fuel cell is operated at temperatures well above 50 °C. Diffusion of
product water from the cathode to the anode side is not sufficient
to compensate losses that are caused by drying processes occurring at
the anode side. Based on our results, the application of a simplified principle in
terms of PEFC without GDL would make rather sense only on the
anode side. The described negative effects on the cathode side as well as a durability that is limited due to catalyst layer degradation,
which mainly affects the cathode side, make a PEFC concept with- out GDL on the cathode side rather unattractive. For the anode side
it might be worth to further investigate a possible application for
the fuel cell industry, but the cost saved by an easier assembly pro-
cess have to be balanced with the technical and economical losses
of such a fuel cell while operating.

5. Conclusions

Performing neutron radiography on an operating micro polymer
electrolyte fuel cell without gas diffusion layers allows insights into
limitations that are caused by liquid water distribution. The micro
flow field structure has to be optimized for the pressure drop and
bypassing effects to ensure that no water accumulation in particu-
lar channels, especially on the cathode side, takes place. Due to the
missing GDL, the membrane material creeps under the clamping
pressure applied to the fuel cell. The excess of creeping depends on
the respective membrane type resulting in mechanical stress affect-
ing the catalyst layer. This mechanical stress can cause rupturing of
the catalyst layer, resulting in a hydrophilic ionomer surface being
exposed to the micro-channels. As a consequence, a water layer is
formed on the cathode side at high current densities, which causes
mass transport limitations. This results in a limiting current density,
which is specific for each membrane type employed. In contrast, the
direct exposure of the anode catalyst layer to the gas flows results
in a strong influence of cell temperature and gas humidification
parameters. For operation temperatures of and above 70 °C, dry-
ing effects have to be counterbalanced by super-saturation of the
hydrogen flow. Decreasing the operation temperature is an alterna-
tive way to compensate these losses because of a less pronounced
drying effect. Finally, we could show that sufficient equilibration
time is necessary to record representative current–voltage curves.

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