Elucidation of Fluid Streamlining in Multi-Layered Porous Transport Layers for Polymer Electrolyte Water Electrolyzers by Operando Neutron Radiography

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Developments of the porous transport layers (PTLs) in recent years resulted in significant performance improvements in polymer electrolyte water electrolyzers (PEWEs). One of the milestones of the material design was the integration of a microporous layer (MPL) on sintered titanium PTLs. Utilizing high-resolution neutron imaging, the water and gas distribution in the multi-layered porous transport media (ML-PTL) was probed at various current densities (up to 4 A cm$^{-2}$) and pressure conditions up to 8 bar, using a series of four materials, differing in MPL morphology. The water and gas distribution measured is greatly affected by the presence of an MPL. While in the bulk of the PTL, the gas accumulation is increased in the presence of an MPL, in the MPL itself more water is retained. The finer the MPL structure, the higher the liquid saturation. It is observed that the two-phase flow in the MPL has minor influence on the performance of the cell even though the gas accumulation at the CL interface is greatly reduced. The improvements, therefore, appear to be related to the CL and MPL interaction on sub-micron scale and microstructure effect on catalyst area utilization.

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The global energy system is slowly adapting to the new green policies as the renewable sources of energy are becoming a baseload supply. 1 In Germany, already over 50% of electricity production is coming from renewables and it is projected that with this time value this will further increase. 2 Unfortunately, the output of the environment-friendly energy technologies (e.g., solar and wind) is highly dependent on weather conditions significantly challenging the grid regulation. 3 Moreover, there is a seasonal trend in the electricity production adding another degree of complexity to the problem. There is a fraction of potentially available energy, which is not utilized due to grid balancing and curtailment. 4,5 By introducing energy, storage and sector coupling on a large-scale these losses can be minimized. One of the key vectors that can enable long-term storage to flatten peaks and valleys between winter and summer seasons is hydrogen. Germany already announced in its national energy strategy to ramp up hydrogen production capacity to 5 GW by 2030 what translates into 1000 commercial average size electrolysis plants. Polymer electrolyte water electrolyzers (PEWE) 6,7,8 are suitable for this role, as this technology is capable of operating under variable loads, produces high purity gas and does not need lengthy start-up procedures. 6-8 PEWE has been identified as one of the key players in the upcoming energy system transition, which will be responsible for both large-scale and decentralized energy storage after achieving market penetration. 9

According to a report published by National Renewable Energy Laboratory (NREL) on PEWE manufacturing costs, 10 the biggest contributors to the MW size stack costs are, listing from the most expensive one: catalyst coated membranes (CCMs) ∼40%, porous transport layers (PTLs) ∼20% and bipolar plates ∼15%. The same studies show, however, that almost two-thirds of the plant cost, stems from the balance of plant (BOP) where the automation and other electric parts are responsible for almost half of the amount. Finally, the cost of hydrogen will also be influenced by electricity prices since the electrolysis process is very power-intensive. Nowadays, technology is still struggling to enter the market with the economies of scales. With further process and design improvements, PEWE has a great potential of becoming a cornerstone of energy storage and solution to sustainable green energy model. 7,11,12

To make PEWE technology more accessible and economically feasible, academic as well as industrial objectives focus on efficiency improvement, 13,14 long-term durability 15,16 and material cost reduction. 17,18 One of the areas on which intensive work is being done is the reduction or replacement of precious group metals (PGMs) in the catalyst layers (CL) 19 and ionomer optimisation. 20-29 These studies focus on obtaining stable and efficient CCMs, potentially reducing the cost of the most expensive PEWE component. Employing non-PGM catalyst layers has proven to be extremely challenging. The biggest issue is the stability of the catalyst on the anode side, where the combination of high potential, acidic environment and oxygen partial pressure make harsh conditions. Even when state-of-the-art catalysts such as Ir based oxides 30,31 are employed, degradation phenomena can be observed. 32

Another aspect in which the CCMs are being refined is the membrane type and thickness. The ohmic losses stemming from the membrane are dominant in overall performance breakdown 37,33 which is why developments in this direction are of great importance. Using thinner membranes decreases the resistance of the cell but on the other hand, has a negative impact on the gas crossover and durability. Investigations of the gas permeation, mitigation and optimal operating conditions have been a topic of various studies 21,34-37 and further developments in those areas might bring significant gains in performance. Assuming that mitigation strategies will enable safe operation of the stacks assembled with thin membranes, improved PTLs with a smoother surface will have to be used in order to support the CCMs under elevated pressures and prevent mechanical damage.

Significant effort is directed towards advancements in PTLs of various structures 38-44 amongst which sintered titanium PTLs have been in the spotlight in recent years due to their superior performance and mechanical integrity. PTLs are key components facilitating educts and product supply as well as thermal and electrical conductivity between the catalyst layer and flow fields. All three main electrochemical losses, associated with mass transport, kinetics and ohmic
overvoltage, are affected by the PTL microstructure.33,45–47 Moreover, the mechanical interaction between the ductile components of the catalyst layer and membrane is dictated by PTL’s surface properties.45 It has been shown using various experimental techniques for single-layer materials, capillary pressure-driven gas transport occurs in the PTLs.8–10 The results conclude that there is no significant difference in the water and gas distribution in the PTLs for a broad range of current densities from 0.005 A cm−2 to 2 A cm−2 and pressures from ambient to 8 bar.50 Novel types of sintered titanium PTLs feature a microporous layer (MPL), which not only reduces the strain on the CCM, enabling the usage of the ultrathin membrane such as Nafion HP (~20 μm) but also increase the performance of a cell.14 In the field of PEMFC the MPL on top of the GDL has already been extensively investigated, and some of the findings could possibly be translated to the PEWEs.52

The functionalty of the PTL is however much different and the impact of the porous structures in electrolyzers is yet unclear which is why it is important to take utmost care when comparing GDLs to PTLs. Improvements in the overall efficiency were also obtained by PTL surface modifications based on coatings.38,64 In order to unveil the origins of the performance increase and gain fundamental insight into the possible influence of the two-phase flow on cell efficiency, a systematic matrix of 4 PTLs with almost identical porosities is investigated. The samples consisted of three multilayer materials with MPLs (ML-PTLs) with different pore sizes of the MPL layer and one single layer material. The last single layer (SL) material was identical to the layers on which the MPLs were integrated. This work, therefore, links directly to the results of Schuler et al.35 in which the performance and material properties of the same PTL structures were thoroughly analysed. The objective of this work is to shed light on the unknown water and gas distributions in ML-PTLs using neutron imaging for a range of current densities and operating pressures up to 8 bar. Finally, the novel insights and advantages of these materials with respect to development, stability and origin of gas pathways are discussed.

Experimental

Neutron imaging and beamline.—The experiments were carried out at the NEUTRA38 thermal neutrons beamline of the SINQ facility of the Paul Scherrer Institute. An anisotropic tilted detector setup was used to push the resolution limits in the direction of interest which resulted in a horizontal pixel size of 6 μm (vertical size: 30 μm) and a field of view of 5 mm (horizontal) × 60 mm (vertical). More information on the tilted detector and its working principle can be found elsewhere.39 The exposure time of a single image was set to 20 s and for each of the measurement points including reference conditions, 60 images were collected. Each of the measurement series was finally averaged together what resulted in total integration times of circa 20 min. A dry cell reference image was used to isolate the signal coming from the water in the PTLs. The images were corrected for scattered neutrons contribution. The scattering background was computed by recording an image with a so-called black body (BB), which is nothing else than a grid of dots which stop the beam, placed in front of the cell. The BB was specially designed for the tilted detector setup for the dots to appear circular. 1.2 mm long and 0.2 mm wide, elliptic holes were made in 1 mm thick aluminum plate by water jet. The holes were then filled with the mixture of gadolinium powder and resin what provided sufficient neutron attenuation. Finally, a 5 × 7 grid was visible in the field of view what is a sufficient amount to apply interpolation function and generate the scattering background (SBKG) image. More information on SBKG correction can be found in the following publication.60 Moreover, detailed information on image processing can be found in previous group publications.50,60,61

The quantification of the images was performed by applying Beer-Lamberts law of attenuation according to:

\[
I = I_0 \cdot e^{\Sigma_i \delta_i} \rightarrow \log \left( \frac{I}{I_0} \right) = \Sigma_i \cdot \delta_i \tag{1}
\]

Where \(I_0\) represents the incident neutron beam flux, \(I\) the attenuated neutron beam flux, \(\delta_i\) the attenuating material thickness of component \(i\) and \(\Sigma_i\) the neutron beam linear attenuation coefficient of the component \(i\).

In this study, the water thickness \(\delta_{H2O}\) ranged between the minimum of 0 mm (fully gas saturated) to ∼3.5–4.4 mm (fully water-saturated/max porosity of PTL bulk filled by water).

Electrochemical cell and test bench.—The cell was assembled with a commercial CCM (Greenerity) based on a Nafion N117 membrane (177 μm thick in dry state conditions), Iridium oxide as anode catalyst (2 mg cm−2), and Pt supported on carbon as a cathode catalyst (1 mg cm−2). This CCM with an active area of 1 cm² was supplied by Greenerity (Hanau, Germany). The cell design was optimized for neutron imaging experiments while maintaining enough versatility to be able to easily adapt to new experimental conditions. Gold-coated stainless steel flow fields ensured that there was no corrosion coming from the cell components. Stainless steel endplates and aluminum compression plates ensured uniform compression of the membrane electrode assembly (MEA). Rod heaters realized the heating of the body of the cell and kept it at a constant temperature of 50 °C. The anode and the cathode side were controlled separately to ensure even temperature distribution across the electrolyzer cross-section. The feed water temperature was controlled by a reservoir heating system and 5 m long heating pipes leading to and from the cell to the test bench. In order to ensure safe operation over long periods, the cathode compartment was ventilated with dry nitrogen gas at the flow rate of 500 ml min−1. The water flow rate was kept at a constant rate of 85 ml min−1 during all experiments. Gold-coated stainless steel flow fields with five straight channels of 1 mm × 1 mm cross-section separated from each other by 1 mm land were used.

Samples and preparation.—Before assembling, each of the CCMs was immersed in ultrapure water (resistance of 18 MΩ cm) for at least 8 h to ensure complete hydration before experiments since lengthy conditioning procedures were not possible during a given amount of beam time. The cells were always assembled in a symmetrical configuration, meaning that the same type of PTL was used on both sides. The samples comprised a single layer (SL) material without MPL and

| Sample | Support layer | MPL |
|--------|---------------|-----|
|        | Thickness (μm) | Pore size (μm) | Porosity (%) | Thickness (μm) | Pore size (μm) | Porosity (%) |
| SL-PTL | 6 μm ML-PTL   | 976 ± 20        | 27.2 ± 1.8 | 34.1 | 220 ± 44        | 6.2 ± 0.2 | 39.8 |
|        | 11 μm ML-MPL  | 268 ± 21        | 11.2 ± 0.2 | 35.2 | 275 ± 40        | 13.2 ± 0.2 | 33.9 |
The first cell to be assembled was the one with a single layer only. For this first sample, a different sequence of pressure values was used than for the subsequent samples due to technical issues, which limited the operating pressure to 8 bars. The pressure lineup for SL-PTL and ML-PTL samples is depicted in Table II.

After the assembly of the cell and integration in the test bench, reference images were taken. Subsequently, the cell was operated and the polarization curves were measured. When assured that the desired pressure was reached and that the cathode PTL was completely dry, the protocol controlling the current density was started. The sequence looped until all of the pressure conditions were measured. Finally before switching to another sample, the high current density point of 4000 mA cm$^{-2}$ was recorded.

### Results and Discussion

The results provide insights on the gas pathways in single and multilayer PTLs by extracting water saturation thicknesses. In the first part, the impact of current density is temporally and spatially resolved the bulk of the MPL, support layer and flow field. Cathodic water removal as a function of pressure is investigated. Subsequently, the relation of PTL microstructure and performance as well as underlying processes are discussed.

**Gas saturation and pathway stability in MPL, PTL and FF domains.**—The images do not contain information that stems from the cell body and CCM due to image processing (only change in water content is indicated) which is why the authors decided to present areas of interest on the radiograms to best guide the reader’s eye. The anode PTL of each of the samples has been divided into three areas, as shown in Fig. 1, which will be referred to further on. Please note that for the SL-PTL sample, there is no MPL but for the sake of consistency, an equivalent area as used for MPL characterization was employed at the interface of PTL and CCM. Later on, referred to as MPL region. It is also important to note that the zero distance mark on the axis is located at the very center of the CCM in order to easily navigate between the cathode (negative numbers) and anode (positive numbers).

Figure 2 presents processed sets of images of chosen data points ranging from kinetic through intermediate to high current density regimes. As already mentioned in the experimental chapter, the $x$ and $y$ dimensions have different scales due to the anisotropic imaging setup with 1:5.7 ratio.

With an optical examination of the images, one can clearly see the separation of each of the anode side PTL areas in water thickness. This effect is most pronounced for the 6 μm ML-PTL sample which is exhibited at the bottom section of Fig. 3. This is already a qualitative indication for significant changes in water gas distribution between SL-PTLs and ML-PTLs.

To gain further insight, the quantitative analysis of the radiograms was performed. The water saturation was determined in different areas and plotted for the entire measurement series.

The analysis of the water thickness in the anode PTLs over the entire measurement duration provides information on the fluid transport mechanisms, which are possibly influenced by transient processes forced by factors other than the change of electrolyzer cell operation parameters (e.g. pressure hysteresis startup and shutdown, gas network pathway alternation). It is necessary to inspect the overall trend in water and gas distribution in the PTLs before an in-depth analysis of every single part to avoid drawing misleading conclusions. Figure 3 presents the average water thickness within the three segments (as defined in Fig. 1) of the PTLs in time for all performed experiments. Holding times of 20 min per galvanostatic step ensured quasi-steady-state operation and stable pathways.

The overall trend shows stable/comparable water thickness values independent of current density at given operational conditions. As shown in Fig. 3, the cell assembled with a single layer only material displayed water and gas distribution independent from current density as well as pressure and point in time throughout the

### Table II. Experimental parameters list and order for each measurement series.

| Series | SL-PTL | ML-PTL |
|--------|--------|--------|
| 1      | 1      | 1      |
| 2      | 5      | 5      |
| 3      | 10     | 8      |
| 4      | 8      | 8      |
| 5      | 8      | 5      |
| 6      | 5      | 1      |
| 7      | 1      | 5      |
| 8      | 1      | 8      |

Three types of ML-PTLs. The basic parameters of the samples are listed in Table I.

Further information and detailed material analysis have been shown in a previous publication. The PTLs were treated in an ultra-sonication bath for 4 h before the assembly in order to extract residual gas from the pore network and guarantee complete ultra-sonication.

Experiment outline.—The experiment consisted of 4 consecutive cell assemblies for which a defined measurement protocol was carried out. Each protocol consisted of 8 measurement series which varied by system pressure. The current density changes within each series were strictly automatized using the sequence whereas the operating pressure setting was done manually. Each series consisted of current densities from 0.01 A cm$^{-2}$, to 2 A cm$^{-2}$ and the holding time was 20 min (for details please see Fig. S2 available online at stacks.iop.org/JES/168/014505/mmedia). After the series was completed an additional point at the current density of 4 A cm$^{-2}$ was measured. The flow chart presenting how the measurements were done and in what order is shown on Fig. S1.
experiment. All three segments across the total PTL thickness kept rather stable water thickness levels with a clear gradient across the PTL thickness. These results align well with our previous study and were expected for this type of single-layer materials as the SL-PTL morphology is comparable to the sintered T10 PTLs manufactured by GKN.

Water thickness of ML-PTLs with 11 μm and 13 μm MPLs show no significant impact on current density dependence at given operating conditions. Variation in operational pressure reveals also no visible trends over the entire measurement duration. The 13 μm ML-PTL and 11 μm ML-PTL bulk show a slight decrease in water thickness over the measurement series. In the absence of clearly identifiable steps related to either, the pressure of current density changes, the different water distributions cannot be identified as an effect of these parameter variations. The data rather suggest that they are the result of a slow stabilization of the water/gas distribution in time. The water thickness levels for the two samples are almost identical and their average values are similar for each presented PTL segment.

The last sample, 6 μm ML-PTL, shows even higher stability during the experiment duration similar to the SL-PTL. Here, a clear separation between MPL and SL is visible by comparing the respective water thicknesses.

Interestingly, for all samples, the first measurement series appears as a slight outlier and only after elevating pressure, a stable water saturation level in the PTL is reached. This does not include the MPL section of the PTL however; here the conditions are strictly defined and stable regardless of time and operating conditions. This result suggests that in order to establish stable pathways in the PTLs, a conditioning step is necessary.

Independent of the operational parameters, a peak of water thickness in the domain next to the catalyst layer is observed in the ML-PTL samples. In the ML-PTL bulk, water saturation levels decrease as compared to the SL-MPL. This phenomenon is discussed in more detail in the next section.

In the area close to the fields, there are significant differences between the samples stemming from the much different gas accumulation levels at the MPL and support layer interface. It also has to be marked that the SL and ML-PTLs are of different thicknesses what will also have a significant impact on the results.

**Impact of PTL microstructure on saturation profiles.**—Comparison between the samples is shown in Fig. 4, which contains the profiles of water thickness across the PTLs for three current densities (0.1 A cm⁻², 2 A cm⁻², 4 A cm⁻²). MPL regions are marked in yellow colors. Please keep in mind that the thickness of the MPL for each sample was slightly different, as seen in Table I.

The resolution in the magnified direction (across the PTL) was 20 μm and in the other direction 150 μm. The porosity of the samples is roughly 36% which means that for our experiment 1 mm of water would translate into ~28% water saturation of the PTL. The absolute water thickness calculation is highly dependent on the scattering background that was measured carefully. We assumed a conservative background measurement error value of 1% of the open beam, what would translate into 0.2 mm water thickness deviation. First of the two current densities are under differential pressure conditions (anode 1 bar and cathode 8 bar) and the last one with balanced ambient pressure.

Focusing on the anode, the presence of the MPL visibly affects the shape of the water thickness profile across the PTL thickness. For
Figure 3. Overview of water thickness in time in respective PTL bulk and FF areas during the entire experiment duration.

Figure 4. Comparison of water thickness profiles across the PTL between all samples for differential pressure conditions (anode 1 bar, cathode 8 bar) for low and intermediate current densities (0.1 A cm\(^{-2}\), 2 A cm\(^{-2}\)) and balanced ambient conditions for 4 A cm\(^{-2}\). MPL regions are highlighted in orange.

* SL sample is thinner than SL-MPLs
SL-PTLs an almost linear water thickness profile establishes between the catalyst layer and flow field as observed in previous studies for single layer PTLs.\textsuperscript{50,51} For ML-PTLs, three defined regions of water saturation levels can be identified.

Region 1 represents the pore space between the CL and the support layer (MPL thickness $\sim 250 \mu$m). The smaller the pore sizes of the MPL, the lower is the gas accumulation near to the catalyst layer. This behavior correlates well with the observation that the flow characteristics in the PTLs are capillary force driven.\textsuperscript{50} On the other hand, open-pore volume in the MPLs and support layer, feature similar values. There is an increased amount of water in the MPL even at the OCV state. Considering that all of the pores were completely filled with water there should be little to none difference in water thicknesses between the samples when there is no gas production. The smaller pore sizes which feature high capillary pressure seem to be more favorable for water filling. Finer morphology would therefore elevate water saturation in the pore network and improve water accessibility to the active sites.

Region 2 captures another eye-catching phenomenon, the rapid transition of high water saturation in the MPL to lower values in the support layer. For ML-PTLs, a peak of water saturation is located close to the MPL, the lower is the gas accumulation near to the catalyst layer. This trend can be observed where CL materials feature a rather linear increase in water thickness between CL and FF. Considering that all of the pores were completely filled with water there should be little to none difference in water thicknesses between the samples when there is no gas production. The smaller pore sizes which feature high capillary pressure seem to be more favorable for water filling. Finer morphology would therefore elevate water saturation in the pore network and improve water accessibility to the active sites.

Region 3 is the support layer bulk domain (support layer and SL-PTL thickness $\sim 1000 \mu$m). The water thickness profile in the support layer features similar characteristics for ML- and SL-PTLs. An almost linear increase in saturation is observed. Though the offset value depends on MPL pore size. The smaller the MPL pores, the higher the gas saturation at the interface between MPL and support layer. Interestingly, all support layers of the ML-PTLs show significantly lower water contents compared to the SL-PTL, considering that the support layer and SL-PTL feature identical morphologies. Moreover, when focusing on the low current density operation ($< 0.5 \text{ A cm}^{-2}$), elevated pressure leads to faster water accumulation, probably due to slower evaporation. What is important to be considered is the fact that during all experiments the cathode was being purged with dry N\textsubscript{2} gas, which definitely affected how fast the PTL was drying. These boundary conditions lead to diffusion-limited evaporation of water transport in the PTLs and need to be compared thoughtfully with a normal operating scenario in which water accumulation dynamics can be different. Besides, a potential impact of water removal on cell and Faradaic efficiency cannot be discerned and is of interest to investigate in future studies. One of the studies that could benefit from these results is related to in situ regeneration of the PEWE cells using the cathodic compartment, which highly relies on CCM/PTL conditions.

Cathodic water removal as a function of pressure.— Measurement of water and gas distribution in the cathode PTL was realized by drying the PTLs before each measurement run and ensuring that the current density protocol was the same for each case. This way it was possible to make a comparison between the samples by analyzing the water thickness in time. The average water thickness in the cathode PTLs with time under various conditions is presented in Fig. 5. The 11 and 13 $\mu$m ML-PTLs feature similar trends as the SL-PTL. The 6 $\mu$m MPL sample exhibits significantly lower average water accumulation, consistent with the anode side.

![Figure 5](image)

Figure 5. Average water thickness in the whole cathode PTL during a single series under various pressure conditions. Jumps indicate current density increases, in the order as shown in Fig. S2.

Here, however, the MPL does not feature excessive water buildup what supports the theory that the water profiles are governed by the material morphology. Moreover, when focusing on the low current density operation ($< 0.5 \text{ A cm}^{-2}$) (<80 min), elevated pressure leads to faster water accumulation, probably due to slower evaporation. The impact of the MPL on the cell performance is not as clear as in the study by Schuler et al.,\textsuperscript{14} the results in terms of water and gas distribution are still valid. The argument that supports this theory is...
that there is a significant performance difference between 13 μm and 11 μm ML-PTLs, but observe very similar water and gas distribution what was to be expected from those very similar materials. Moreover, these underperforming cells were still fully operable in the whole parameter range at reasonable voltages. We attribute the performance spread to differences in compression and/or the absence of a long conditioning process due to time constraints and cell reassembly during the beam time. Finally, we believe that the general performance of the cells is good enough to say with confidence that they are reproducible in terms of water/gas distribution.

In the previous study, significant improvements are obtained when using samples with MPLs. Origin of the performance increase has been identified to stem from increased CL utilization due to finer surface of the ML-PTL. Also, mass transport losses are shown to be attributed to catalyst layer transport resistance and not to fluid transport in the bulk of the SL or MPL.

The novel insights on fluid transport of this study are complementary and can further elucidate the overall role of fluid transport in PTLs. While Schuler et al. did show that improvements brought by the MPL are similar for all samples (6 μm, 11 μm and 13 μm MPLs), this study shows that the gas accumulation at the CL-MPL interface of the 6 μm ML-PTL is lowered in a more pronounced way. This result further indicates that the performance improvement from PTL with MPLs is not primarily driven by the prevention of gas accumulation near the catalyst layer—in which case the 6 μm MPL would have been outperforming the remaining PTL types.

The results suggest that processes related to catalyst, layer and CL/PTL interface are an important factor for the cell efficiency and that the fluid transport in the bulk material for this type of PTL materials is not a limitation to the cell performance. In order to gain further understanding, investigations on the CL level are of high interest. 3D resolved imaging of fluid transport in Ti-based ML-PTL is indispensable to improve further understanding of at the transition region and the interface near the catalyst layer in future studies.

The relevant implication of our results focuses on possible cost reductions for PTL manufacturing. We identified that the presence of an MPL while reducing the gas accumulation at the CL interface results in an increased gas saturation in the bulk of the PTL. The fact that this unfavourable water/gas balance in the PTL bulk has no negative impact on performance can guide the design of PTL products with reduced feedstock costs or advanced manufacturing methods.

**The underlying mechanism of water and gas streamlining in multilayer PTLs.**—The fine structures of the MPL with its smaller pore sizes provide improved water filling properties as compared to SL-PTLs. This is due to increased capillary pressure for the gas phase and therefore higher water content. Since fluid transport is based on a capillary fingering flow regime, the gas produced at the surface of the catalyst layer is injected in the pores of the microporous layers and proceeds pore by pore following the path of lowest resistance according to capillary fingering mode. Over the thickness of the MPL, the multiple injection points/gas pathways merge and the liquid saturation increases. The gas will immediately fill large pores upon leaving fine pore areas what we believe is the explanation behind the water thickness minimum at the MPL—SL interface (see Fig. 7).

The ML-PTL can be considered as a cascade of two such elements, streamlining gas paths based on identical path merging processes. At the interface between MPL and support layer, the gas accumulation increases due to the injection of gas from multiple small pores in a single big pore. The result implies that all the large SL pores at the MPL-SL interface will be highly occupied by gas at first before merging again and reducing gas saturation. This explains the sharp increase in gas accumulation at the MPL/SL interface. In the support layer gas and water undergoes a second streamlining with merging of several pathways in less gas-filled pores across the SL thickness. The interfaces between the different layers and in particular pore size ratios appear to be a parameter, which can be used for adjusting saturation profiles across the PTLs.

**Conclusions**

The two-phase flow pattern for four sintered titanium porous transport layers comprised of three multilayer PTLs and one single layer PTL were investigated under elevated pressures up to 8 bar and current densities up to 4 A cm$^{-2}$ by operando neutron imaging. Materials characterized in this study were reported in detail in a previous publication focusing on the material properties and their impact on the cell performance.

The present imaging results show that the presence of a microporous layer strongly affects the fluid transport across the entire anodic PTL. Three regions of interest were identified:

- **Region 1:** The microporous layers lead to lower gas accumulation at the CL interface providing higher accessibility of water to active sites in the catalyst layer.
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