Neutron radiography was applied to image the water-gas distribution in an operating small scale PEM electrolyser, building up on the anisotropic resolution improvements previously developed for fuel cell imaging. The ability of neutrons to image this distribution across porous materials made of titanium was demonstrated for the first time. This paper presents the procedure’s description and limitations, focusing on water content in the porous layers as a function of distance from electrode. A surprising pattern of water content in anodic porous material was observed, which sheds light on the topic of mass transport limitations in PEM electrolyser.

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Imaging.—Imaging was performed at the Paul Scherrer Institute’s SINQ neutron source at the NEUTRA thermal neutron radiography beamline.\textsuperscript{14} The investigated orientation was so-called in-plane setup (neutron beam parallel to membrane), with channels parallel to the beam, anode at the top. Images were taken with exposure time of 10 seconds and averaged over 5 minutes, in order to achieve sufficient signal-to-noise ratios. The cell operating parameters were kept constant for at least 5 minutes prior to imaging start. Images of the dry cell were taken as a reference for the image processing.

Calibration.—Calibration was performed according to the procedure described in Ref. \textsuperscript{15} and the supplementary material; as a result the linear attenuation coefficient value was estimated to be 3.5 cm\textsuperscript{-1}, which is in good agreement with the value previously stated for the NEUTRA beamline.\textsuperscript{9}

Results and Discussion

Radiograms obtained in in-plane configuration (shown in Fig. 2) are the result of averaging over 5 minutes periods. The corresponding electrochemical data is shown in Fig. 3. Both anodic channels (upper borders) and the membrane (horizontal stripe in the middle) do not have sufficient transmittance for reliable analysis. The effective resolution in the vertical direction (across the membrane) was estimated to be approximately 75 µm.

Anodic porous layer.—When no current was applied to the cell, a uniform water distribution was quickly established in the anodic porous layer — this process was too fast to follow with the used exposure time.

After applying a current (the lowest examined current density was 0.1 A cm\textsuperscript{-2}) a new pattern emerged. While in direction parallel-to-membrane the water content was still fairly uniform (a weak rib-channel distribution can be observed at distances lower than 0.1 mm from the flow field), in orthogonal direction it is no longer valid. Instead, a linear gradient was established (Fig. 3), ranging from ca 85\% (at the flow field) to 55\% (at the membrane) of initial water thickness, independent on current density (the highest examined value was 2.5 A cm\textsuperscript{-2}).

Cathodic porous layer.—In the lowest current cases, the measured water thickness is around 0.2 mm, which is visible in Fig. 3. Attributing this to residual water presence is hardly possible, since the area of decreased transmission extends outside the porous layer. A plausible explanation is, that the reference images are biased by stronger background coming from scattering within the detector setup.

General.—We have found the observed patterns to be counter-intuitive. The strong gradient on the anode side indicates a dynamic equilibrium between the transport of water to the electrode and the transport of gas away from the electrode. The fact that this equilibrium is unaffected by the magnitude of the flows over a wide range (from 0.1 A cm\textsuperscript{-2} to 2.5 A cm\textsuperscript{-2}) is surprising. Because the heat production strongly varies with the current density, this also suggests that the thermal gradient does not play a major role in the water/gas distribution. The observed pattern clearly deserves attention as it can lead to the development of models for both transport in porous media and electro-osmotic drag. At all current densities, the water content in the vicinity of the electrode is approximately half of the initial value,
which may suggest, that the water availability at the catalyst surface can be treated only on the microscale, with transport processes through the porous layer being neglected. Close to the channels, the attenuation was hardly distinguishable from the zero-current situation. This shows that water transport through this interface is fast. Contradicting conclusions were drawn in Ref. 3, where the dependence of the overpotential on the water flow rate was observed. These results were however obtained at different range of superficial water velocities, as well as different cell design, which may explain this discrepancy.

Summary
For the first time, neutron radiography was used to measure the water/gas distribution across the structure of an operating PEM electrolysis cell, with a focus of the water content within the porous media made of sintered titanium. The in-plane configuration was shown to give a detailed insight into the processes occurring in the porous media. The observed pattern of water-gas distribution inside the porous media is surprising. The obtained gradient is unaffected by the local reactants’ flow variation over a large range of current densities (0.1 A cm\(^{-2}\) to 2.5 A cm\(^{-2}\)), not showing any sign of water-starvation at high current; although not yet fully understood, these observation are an important starting point for the understanding and modelling of mass transport in PEM electrolyzers.

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