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Effect of ageing of gas diffusion layers on the water distribution in flow field channels of polymer electrolyte membrane fuel cells

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HIGHLIGHTS

- Water droplet size distribution measured by means of neutron imaging.
- Artificial GDL ageing applied to study effects on water distribution and transport.
- Droplet sizes in flow field channels decrease with increasing GDL ageing.
- Water on the GDL surface favourable blocks the gas supply with increasing GDL ageing.

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ABSTRACT

We present a quantitative analysis of the influence of artificial ageing of gas diffusion layers (GDL) on the water distribution and transport in polymer electrolyte membrane fuel cells (PEMFCs) during cell operation. Water droplet size distributions are measured by means of in-operando neutron radiography. We find a strong correlation between droplet size distribution and GDL ageing time: With increasing GDL ageing, water droplet sizes in the flow field channels strongly decrease, indicating an ineffective water transport that leads to a reduced cell performance. This effect can be assigned to water accumulations on the GDL surface that block the gas supply towards the catalyst layer.

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1. Introduction

Fuel cells will play an important role in a future hydrogen economy [1–3]. Because of their high power density and flexible operating range, low-temperature polymer electrolyte membrane fuel cells (PEMFC) are suitable for mobile applications, e.g. in vehicles [4] or to provide combined heat and power in stationary energy systems [5,6]. A decisive factor for the optimal operation of PEMFCs is a balanced water management [7,8].

On the one hand, the membrane of the PEMFC must be sufficiently humidified to be proton conductive, but on the other hand, it must not accumulate too much water since otherwise the pores of the gas diffusion layer (GDL) will be clogged and the supply of the catalyst layer with reactant gases obstructed [3,7,9–12]. During long-term operation, the properties of the GDL fibres change [13,14]. Many published works examine the water transport and distribution with synchrotron X-ray imaging, which is characterised by a high temporal and spatial resolution [9–11]. Others have visualised the water transport with transparent cell materials [15]. In this article, the effect of GDL ageing on the water balance of a fuel cell is shown by neutron imaging [16–21]. The strong attenuation of the neutron beam by hydrogen combined with the

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high transmission through the fuel cell components allows for a high contrast and a quantitative investigation of the water distribution [22–26].

2. Experimental

2.1. Neutron radiography

The imaging instrument CONRAD (**CO**ld **N**eutron **Rad**iography) is designed for materials and structure research [27]. It provides the possibility of creating radiographs using cold neutrons. The neutron beam at CONRAD is formed by a neutron guide and is polychromatic with wavelengths between 2 and 6 Å and a maximum intensity at about 3.0 Å. The measurements of the fuel cells discussed here are performed 10 m behind the exit of the neutron guide. Although this large distance leads to a lower neutron flux density and thus a lower image repetition frequency, i.e. a loss of temporal resolution, it allows for a higher spatial resolution [28].

Behind the sample, a detector system consisting of a scintillator, mirror, lens and CCD camera is placed [28–30]. When the neutrons, which are transmitted and scattered by the sample, hit the scintillator, photons in the visible spectrum are emitted. The scintillator used for the measurements is a lithium fluoride crystal with silver-doped zinc sulfide (6 LiF/ZnS (Ag)).

The photons are projected onto the camera by a mirror/lens combination. The Andor DW436 camera used contains a 16 bit chip with (2048 × 2048) pixels, each of a size of 13.5 μm [27]. The CCD sensor is continuously cooled to below −50 °C to keep thermal noise as low as possible. With the optics used an imaging field of view of (108 × 108) mm^2 with a pixel size of 53 μm is achieved. Each radiographic projection is acquired with an exposure time of 16 s.

2.2. Fuel cell setup

Four single cells with an active area of 10 cm × 10 cm are investigated. For each cell, a differently aged GDL is used (see details below). A GORE® PRIMEA 5761™ catalyst coated membrane is applied. The regulation of the cell temperature is carried out by using a cooling circuit with deuterium oxide (D_2O). Compared to hydrogen, the attenuation coefficient of deuterium is much smaller [31,32]. As a result, D_2O hardly attenuates a neutron beam and can only be seen faintly in radiographs. The flow field of the cooling circuit is embedded in the backside of the bipolar plates and is connected with a secondary water coolant circuit via a heat exchanger. Into the graphite material of the bipolar plates, three ~0.8 mm wide meander-shaped channels are milled, in which the supply gases, hydrogen on the anode and air on the cathode side, flow from top to bottom. Fig. 1 shows the cell and the flow field of the cathode side. Note that the anode and cathode flow fields are identical, but start in opposite corners, which means that in the radiographs the curves of the anode channels overlap with the ribs at the cathode side and vice versa. The presented measurements are conducted at a current density of 1 A cm-2 and a cell temperature of 50 °C, while the cell is supplied with gases of 100% relative humidity. Via gas flow controllers the ratio of the supplied and consumed gases is set to four for the anode (H_2) as well as the cathode (air) side. To simulate the effect of chemical ageing on cell performance the GDLs of type Sigracet® SGL 25BC are subjected to accelerated ageing at the ZSW.

2.3. Accelerated ageing process

Degradation effects of fuel cells can usually only be registered after several hundred hours of continuous operation. Therefore, accelerated ageing methods are used to enable their characterization in reasonable times. Using ex-situ ageing ensures that the desired ageing process is isolated from other degradation mechanisms in the fuel cell. The method used here for accelerated ex-situ ageing of the GDL material is conducted with hydrogen peroxide (H_2O_2). The gas diffusion layers are exposed to a solution of 30% H_2O_2 (diluted with water) at 90 °C for several hours. After this, the GDL is washed for 1 h in distilled water to remove the H_2O_2 residues and then dried at 80 °C. For our study, GDLs of type SGL 25BC were chosen. Both the cathodic and the anodic GDLs of the four test cells were subjected to accelerated ageing by H_2O_2 treatment of the GDLs for 0 h, 4 h, 8 h and 24 h.

2.4. Analysis of water droplets in the channel

As one main purpose of this work is the analysis of water droplets in the channel system the procedures to characterize them are described in this chapter. The thickness of the water layer through which the beam locally passes is calculated by Lambert–Beer's law [31,33,34]. By setting a grey value as a threshold, individual water accumulations above a certain thickness are extracted and size and number of them are determined (The effect of different parameters is shown in Fig. 2). The threshold for this droplet selection must be chosen appropriately. If it is set too large, a high proportion of the water accumulation shown in the radiographs no longer appears in the resulting images (blue arrows in Fig. 2 A). If the selected threshold is too low it will not be possible to distinguish single water droplets from one another (see red arrows). As a good compromise, a threshold value of 200 μm has been used for the radiographs shown here. This threshold is also sufficiently high to make GDL water accumulations under the ribs invisible.

The number and size of water accumulations in the channels are determined with the “Analyze Particle” function implemented in the open-source image processing software ImageJ. Artefacts arising due to image noise are removed by setting a minimum lateral size for the selection of water agglomerations. The value for this second criterion is set to 10 pixels (equivalent to 0.028 mm^2). The effect of different settings can be seen in Fig. 2B.

According to the specified minimum thickness and minimum size in pixels, the minimum volume of the selected water droplets for being detected is ~5 nl. Not all the water in the channel fits in these criteria. At the same time also water agglomerations could be selected, which are incidentally overlapping in the projection and reach a thickness of more than 200 μm. However, the method is suitable for qualitative studies of droplet size distributions and the transport behaviour from the GDL into the channel and the comparison of aged and non-aged materials.

3. Results

Ex-situ investigations revealed a decrease in contact angle of water and carbon fibres due to ageing in H_2O_2 solution, which corresponds to a decrease in hydrophobicity [35]. The contact angle of water droplets on the GDL surface decreases from 160° (0 h) to about 98° (24 h H_2O_2 ageing). Correspondingly, the voltage of the test cells decreases. At 1 A cm-2 current density, the voltage obtained drops from 650 mV (0 h) to 350 mV (24 h). Although the water production rate is equal for all four cells, the way the water is moved out of the cell is observed to vary with the duration of ageing.

The volume of each selected droplet is calculated via the lateral size and average thickness. A result of the cell containing the pristine material at a current density of 1 A cm-2 is shown exemplarily in Fig. 3. For each radiograph, the number (upper) and the

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