INTRODUCTION

Fuel cells are part of an important key enabling technology for achieving carbon dioxide free emitting electricity generation and can be used for stationary, mobile and portable applications (Hoogers, 2003; Vielstich et al., 2003). A polymer electrolyte membrane fuel cell (PEMFC) is an electrochemical cell that is fed with hydrogen, which is oxidized at the anode, while oxygen is reduced at the cathode. As a result of the reaction, water will be formed. For that, a well-balanced water management is important to avoid two negative operation situations: Excessive drying of the membrane and flooding of the diffusion media (Carrette et al., 2001; Garche et al., 2009; Vielstich et al., 2003; Wang, 2003). Thus, the cell performance drops. The water produced at the cathode catalyst layer exits the fuel cell either through the cathode flow channels or by back diffusion through the membrane to the anode flow channels. Hence, a well-balanced water management especially under critical operation conditions, such as temperatures below 60°C as well as high currents is an essential condition for optimum power output and long term stability (Gostick et al., 2009; Kitahara et al., 2010; Qi & Kaufman, 2002; Quick et al., 2009). Visualizing liquid water using in-situ imaging during cell operation is a well-established characterization method with many uses (Bazylak, 2009; Bellows et al., 1999; Haußmann et al., 2013; Hinebaugh et al., 2012; Lange et al., 2010; Maier et al., 2012; Manke et al., 2011; Markotter et al., 2012). Several imaging diagnostic techniques have been used to study PEMFCs (Alrwashdeh et al., 2017; Krüger et al., 2009, 2011; Le & Zhou, 2009; Litster et al., 2006; Manke et al., 2008, 2010; Markotter et al., 2012). Among the techniques used for water management studies of PEMFCs are X-ray and synchrotron X-ray imaging (Alrwashdeh et al., 2016a, 2016b, 2017; Hinebaugh et al., 2012). These techniques are able to quantify the dynamic distributions of liquid water during cell operation by measuring the change in beam attenuation at high spatial resolutions.

In this investigation, synchrotron X-ray imaging was used to investigate the water distribution inside newly developed gas diffusion media in polymer electrolyte membrane fuel cells. In-situ radiography was used to reveal the relationship between the structure of the microporous layer (MPL) and the water flow in a newly developed MPL equipped with randomly arranged holes. A strong influence of these holes on the overall water transport was found. This contribution provides a brief overview to some of our recent activities on this research field.

Key Words: Polymer electrolyte membrane fuel cell, Microporous layer, Water distribution, Radiography, Synchrotron X-ray imaging.
In this study, a modified Freudenberg gas diffusion media (GDM) with randomly arranged holes in the microporous layer (MPL) was subjected to synchrotron X-ray imaging to investigate the dynamic liquid water transport behavior. The modified material is compared to unmodified reference material at cell temperatures of 40°C. This work is based on previous work of the authors that was published by Alrwashdeh et al. (2016, 2017).

MATERIALS AND METHODS

Two PEMFCs with active areas of 5.4 cm² and seven parallel vertical flow field channels on both sides were investigated (Alrwashdeh et al., 2016). The first cell having a modified GDM with a gas diffusion layer (GDL) from Freudenberg based on a H1411 fiber substrate, while the second cell contains a reference GDM with a H1410 14 C10 GDL (Alrwashdeh et al., 2016). The modified GDM has a newly developed MPL with randomly distributed holes with diameters ranging up to 30 µm. During the measurement, operating conditions were held with a current density of 1 A/cm² at stoichiometric ratios of 5 on both sides. A cell temperature of 40°C was used in this study. More details can be found in Alrwashdeh et al. (2016) (Fig. 1).

Synchrotron X-ray radiography was used to test a region centered in the middle height of the cell covering an area of ~10% of the total active area (Alink et al., 2013). The measurements were performed at the imaging beamline “BAMline” at the synchrotron electron storage ring Bessy II in Berlin, Germany (Görner et al., 2001). A field of view of 8.8×5.9 mm, and pixel sizes of 2.2×2.2 µm were used (Alrwashdeh et al., 2016). The exposure time for each radiographic projection was 2S and a photon energy of 19 keV was selected, ensuring sufficient transmission through the cell materials while maintaining adequate contrast to water (Alrwashdeh et al., 2016).

RESULTS

At the same operation parameters (1 A/cm², 40°C, stoichiometric ratio 5), the obtained voltage of the reference and modified cells were 500 and 550 mV, respectively. The holes in the MPL might facilitate accumulation of liquid water. The emerging product water then moves through the GDL into the channel, from where droplets are removed continuously by the gas stream. In some cases, water is always transported through the same passage, which leads to droplets originating at the very same positions over and over again (Alrwashdeh et al., 2016).

Few paths are used for water transport according to the radiographic data. Fig. 2A and B show the activity map of the modified Fig. 2A and reference Fig. 2B cells. These activity maps highlight areas with strong temporal fluctuations of the local water amount, which especially applies to the droplets (Alrwashdeh et al., 2016). Three marked droplets shown, act as very active points with a cyclic behaviour (Fig. 2A).

Fig. 2D shows the water volume as a function of time for a selected droplet, see Fig. 2C, which shows a radiographic still of droplet #3. The water droplets periodically built as becomes visible in the graph roughly every 30 seconds, and ends each time in a significant decrease of the measured water volume. The droplet grows at a rate of 0.13 nL/s, as derived from the

![Fig. 1. Schematic drawing of the used radiography setup (A), tomographic slice through a cell with a reference microporous layer (MPL) (B), with a modified MPL (C) and a perpendicular cut shows the distributed holes marked with red arrows (D).](image)
inclining slopes of the graph in Fig. 2D. For droplets #1 and #2 shown in Fig. 2A, the corresponding volume increase rate was 0.22 nL/s and 0.37 nL/s, respectively. These growth rates are related to electrochemically active areas of 0.23, 0.40 and 0.14 mm$^2$ for droplets 1, 2, and 3, respectively (Alrwashdeh et al., 2016).

**DISCUSSION**

The water droplets are transported differently from the GDL through the channel out of the cell which contains the modified GDM. Therefore, it can be concluded that in comparison to the reference material the holes in the modified GDM cause the water to flow only through well-defined pathways as shown in Fig. 2A.

The transport paths leave larger parts of the GDM free of liquid water, i.e. more empty pore space is available for the transport of reaction gases and the supply of the catalyst. The optimized gas supply leads to an improved performance of the cell. Hence, an intentional material modification for an effective water removal from the GDL into the channel is observed (Alrwashdeh et al., 2016).

**CONCLUSIONS**

Transport of liquid water through the GDL and the channel system in a PEMFC was investigated with synchrotron X-ray imaging. Previous studies have shown a strong influence of unintentionally caused cracks in the MPL and perforations of the GDM (Krüger et al., 2011; Markotter et al., 2013; Sasabe et al., 2011). Here, it could be demonstrated that this effect can be exploited with a tailored GDM containing artificial holes in the MPL. The holes in the GDM fill up with liquid water and may provide paths for fast water transport through the
This study can contribute to the optimization of the performance of fuel cells in future.

CONFLICT OF INTEREST

No potential conflict of interest relevant to this article was reported.

REFERENCES

Alink R, et al. (2013) The influence of porous transport layer modifications on the water management in polymer electrolyte membrane fuel cells. J. Power Sources 233, 358-368.

Alrwashdeh S, et al. (2017) Improved performance of polymer electrolyte membrane fuel cells with modified micro porous layer structures. Energy Technol.

Alrwashdeh S, et al. (2016) Investigation of water transport dynamics in polymer electrolyte membrane fuel cells based on high porous micro porous layers. Energy 102, 161-165.

Alrwashdeh S S, et al. (2016) X-ray tomographic investigation of water distribution in polymer electrolyte membrane fuel cells with different gas diffusion media. ECS Trans. 72, 99-106.

Bazylak A (2009) Liquid water visualization in PEM fuel cells: a review. Int. J. Hydrogen Energy 34, 3845-3857.

Bellows R J, et al. (1999) Neutron imaging technique for in situ measurement of water transport gradients within nafion in polymer electrolyte fuel cells. J. Electrochem. Soc. 146, 1099-1103.

Carrette L, Friedrich K A, and Stimming U (2001) Fuel cells–fundamentals and applications. Fuel Cells 1, 5-39.

Garcke J, et al. (2009) Encyclopedia of Electrochemical Power Sources, p. 4538, (Elsevier, Amsterdam).

Gostick J T, et al. (2009) On the role of the microporous layer in PEMFC operation. Electrochem. Commun. 11, 576-579.

Görner W, et al. (2001) BAMline: the first hard X-ray beamline at BESSY II. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 467-468(1), 703-706.

Haußmann J, et al. (2013) Synchrotron radiography and tomography of water transport in perforated gas diffusion media. J. Power Sources 239, 611-622.

Hinebaugh J, Challia P R, and Bazylak A (2012) Accounting for low-frequency synchrotron X-ray beam position fluctuations for dynamic visualizations. J. Synchrotron Radiat. 19, 994-1000.

Hinebaugh J, Lee J, and Bazylak A (2012) Visualizing liquid water evolution in a PEM fuel cell using synchrotron X-ray radiography. J. Electrochem. Soc. 159, F826-F830.

Hoogers G (2003) Fuel Cell Technology Handbook (CRC Press LLC, Boca Raton, FL).

Kitahara T, Konomi T, and Nakajima H (2010) Microporous layer coated gas diffusion layers for enhanced performance of polymer electrolyte fuel cells. J. Power Sources 195, 2202-2211.

Krüger P, et al. (2009) In situ Visualization of Liquid Water Formation and Transport Processes in PEM Fuel Cells Meeting Abstract - Electrochemical Society 902: p. 850.

Krüger P, et al. (2011) Synchrotron X-ray tomography for investigations of water distribution in polymer electrolyte membrane fuel cells. J. Power Sources 196, 5250-5255.

Lange A, et al. (2010) Reconstruction of limited CT data of fuel cell components using DIRECTT. J. Power Sources 196, 5293-5298.

Le A D and Zhou B (2009) Fundamental understanding of liquid water effects on the performance of a PEMC with serpentine-parallel channels. Electrochim. Acta 54, 2137-2154.

Litster S, Sinton D, and Djilali N (2006) Ex situ visualization of liquid water transport in PEM fuel cell gas diffusion layers. J. Power Sources 154, 95-105.

Maier W, et al. (2012) Correlation of synchrotron X-ray radiography and electrochemical impedance spectroscopy for the investigation of HT-PEFCs. J. Electrochem. Soc. 159, F398-F404.

Manke I, et al. (2008) Characterization of water exchange and two-phase flow in porous gas diffusion materials by hydrogen-deuterium contrast neutron radiography. Appl. Phys. Lett. 92, 244101.

Manke I, et al. (2010) In situ synchrotron X-ray radiography investigations of water transport in PEM fuel cells. Fuel Cells. 10, 26-34.

Manke I, et al. (2011) Investigation of water evolution and transport in fuel cells with high resolution synchrotron x-ray radiography. Appl. Phys. Lett. 90, 1-3.

Markotter H, et al. (2012) Neutron tomographic investigations of water distributions in polymer electrolyte membrane fuel cell stacks. J. Power Sources 219, 120-125.

Markotter H, et al. (2012) Visualization of the water distribution in perforated gas diffusion layers by means of synchrotron X-ray radiography. Int. J. Hydrogen Energy 37, 7757-7761.

Markotter H, et al. (2013) Influence of cracks in the microporous layer on the water distribution in a PEM fuel cell investigated by synchrotron radiography. Electrochim. Commun. 34, 22-24.

Qi Z G and Kaufman A (2002) Improvement of water management by a microporous sublayer for PEM fuel cells. J. Power Sources 109, 38-46.

Quick C et al. (2009) Characterization of water transport in gas diffusion media. J. Power Sources 190, 110-120.

Sasabe T, et al. (2011) Soft X-ray visualization of the liquid water transport within the cracks of micro porous layer in PEMFC. Electrochem. Commun. 13, 638-641.

Vielstich W, Lamm A, and Gasteiger H A (2003) Handbook of Fuel Cells–Fundamentals, Technology and Applications, Vol. 3 (John Wiley & Sons, Chichester).

Wang C Y (2003) Two-phase flow and transport, in Handbook of Fuel Cells–Fundamentals, Technology and Applications, eds. In: Vielstich W, Lamm A, and Gasteiger H A, pp. 337-347, (John Wiley & Sons, Chichester).