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Flooding of the diffusion layer in a polymer electrolyte fuel cell: Experimental and modelling analysis

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ABSTRACT

Water management is widely investigated because it affects both the performance and the lifetime of polymer electrolyte fuel cells. Membrane hydration is necessary to ensure the high proton conductivity, but too much water can cause flooding and pore obstruction within the cathode gas diffusion layer and the electrode. Experimental studies prove that the characteristics of the diffusion layer have great influence on water transport; the introduction of a micro-porous layer between the gas diffusion layer and the electrode reduces flooding and stabilizes the performance of the fuel cell, although the reason is not fully explained. A quantitative method to characterize water transport through the diffusion layers was proposed in our previous work, and the present work aims to further understand the flooding phenomenon and the role of the micro-porous layer. The improved experimental setup and methodology allow an accurate and reliable evaluation of water transport through the diffusion layer in a wide range of operating conditions. The proposed 1D + 1D model faithfully reproduces the experimental data adopting effective diffusivity values in agreement with literature. The presented experimental and modelling analysis allows us to evaluate the influence of pore obstruction on the effective diffusivity, the overall transport coefficient and water flow through the diffusion layer, elucidating the effect of the micro-porous layer on fuel cell performance and operation stability.

1. Introduction

Polymer electrolyte fuel cells (PEFC) are very promising for CHP and automotive applications because of their high efficiency, very low emissions and modularity. However, they are still affected by technical problems that restrict their lifetime and performance. Water management in PEFC is one of the most investigated issues [1,2]. Indeed, polymer membrane hydration is necessary to ensure high proton conductivity, but too much water can cause flooding within the cathode gas diffusion layer (GDL) and the electrode. The flooding in the GDL or the electrode causes pore obstruction due to water condensation, which hinders oxygen transport from the distributor to the catalyst active sites and lowers the performance and the lifetime of the fuel cell [3]. Even with stack optimization, flooding can still occur during the real operation because of the variability in the operating conditions, which gives rise to transients in the water production and transport phenomena. In fact, water transport in PEFC is a result of several contributions such as electro-osmotic drag, diffusive and convective phenomena with phase transition and local water production. Reactant humidification is necessary to avoid membrane dehydration, and it has to be optimized considering the water transport properties of the porous layers. Experimental studies confirm that GDL characteristics have great influence on these phenomena [4]; the introduction of a micro-porous layer (MPL) between the GDL and the electrode reduces the flooding effect and stabilizes the fuel cell performance although the reason is not fully explained [5–7]. Despite its arguable relevance, GDL flooding is not exhaustively investigated in the literature; actually, no common methodologies to compare the different properties of GDL water transport are available. In most experimental studies, these properties are investigated by characterising two physical indicators during the fuel cell operation [8–13]: the overall performance and the water flow at the cathode outlet. Under these conditions, the interplay of different phenomena complicates the interpretation of the results. In the literature, the direct investigations of the water transport and the flooding mechanism in GDL are generally more qualitative than quantitative. For example, they are investigated through infrared thermography during the fuel cell operation, showing that the flooded area is warmer than the not flooded one [14]. In some studies, the water dynamics inside the fuel cell are characterised

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Nomenclature

C	local concentration [mol m^{-3}]
$\hat{C}$	local concentration in the gas phase [mol m^{-3}]
v	velocity of the flow [m s^{-1}]
P	pressure [Pa]
x	axial direction [m]
L	channel length [m]
a	channel width and height [m]
T	GDL temperature [K]
R	universal gas constant [J mol^{-1} K^{-1}]
t	thickness [m]
D	diffusivity [m^2 s^{-1}]
k	local water transport coefficient [m s^{-1}]
h	convective mass transport coefficient [m s^{-1}]
FC	local flooding coefficient
F	2D factor
$\dot{N}$	molar flow [mol s^{-1}]
$\dot{V}$	volumetric flow [m^3 s^{-1}]

Subscripts

2p	relative to a two-phase flow
d	relative to a dry flow
h	relative to a humid flow
H_2O	relative to water
air	relative to air
eff	effective
overall	including possible pore obstruction
sat	relative to saturation
in	at the inlet
out	at the outlet

superscripts

h	relative to a humid flow
d	relative to a dry flow
MPL	relative to MPL
GDL	relative to GDL
GDL + MPL	relative to GDL with MPL
tot	total
diff	diffusive
perm	permeative

using optical techniques such as neutron imaging [15,16] and X-ray imaging [17,18], providing evidence for the water cluster formation and the preferential liquid pathways in the porous material.

In [19], Casalegno et al. proposed a quantitative method to characterise the water transport through the gas diffusion layers by introducing a global water transport coefficient that allows a comparison of different GDLs. The porous media were analysed in realistically simulated operating conditions, which were obtained by supplying the two GDL faces with humid and dry airflows. The measurements showed that a reduction in water transport occurred when there was a significant amount of liquid water, which could be attributed to flooding, and that the MPL had a beneficial influence on the flooding effect.

This work aims to further understand the flooding phenomenon and the MPL role through the following goals:

- improving the experimental setup to achieve a wider range of operating conditions;
- developing a 1D + 1D model, which is validated by the experimental results.

2. Experimental methodology

2.1. Experimental setup

The experimental approach, as reported in detail in [19], consists of supplying the two GDL faces with a humid air flow and a dry air flow in a co-current configuration so that the water flows from the humid side to the dry side by diffusion. The pressure difference between the two flows has to be minimized to make permeation negligible compared to diffusion. Fig. 1 shows schematics of the experimental setup. The water flux through the dry side of the GDL is calculated as follows:

$$\dot{N}_{H_2O}^{GDL} = \dot{V}_{out}^{d} \cdot C_{H_2O,out}^{d} - \dot{V}_{in}^{d} \cdot C_{H_2O,in}^{d} \quad (1)$$

The experimental setup is improved with the following design:

- a more effective evaporator, which allows a more uniform and stable water evaporation in dry air, to decrease the variability and the uncertainty of the relative humidity measurement;
- an improved temperature control system, which permits a more accurate temperature control for different components (the typical deviation is less than 0.1 °C) and an extension of the maximum allowed temperature of the humidity sensors (up to 110 °C).

These modifications allow a significant extension of the operating condition range: the maximum inlet water concentration is increased from 11.5 mol m^{-3} to 15 mol m^{-3}, permitting the investigation of more intensive flooding conditions.

The gas diffusion layer, of which the surface area that is exposed to the fluxes is 4.2 cm × 4.2 cm, is contained between two graphite distributors, where the channels for the humid and the dry airflows have been grooved (both distributors have a triple serpentine channel with a square section: depth 0.8 mm, width 0.8 mm, length 700 mm). The graphite distributors are held together with two stainless steel plates using 8 retaining bolts that are closed with a controlled torque of 12 ± 0.5 N m. The thickness of the compressed GDL is maintained constant at approximately 330 μm, to adopt the appropriate gaskets. A slot in one of the steel plates accommodates a calibrated thermocouple, which is connected with a temperature controller and a data acquisition system. Two electrical heaters, which are connected to the temperature controller, are placed within the steel plates to fully control the temperature of the assembly. Because the heat capacity of the plates is much greater than that of the graphite distributors and the GDL, high temperature stability is attained. The rates of the airflows are controlled and measured by two calibrated flow controllers. The air humidification is obtained by adding bi-distilled water to the air stream with a precise peristaltic pump. Air pressure, temperature and humidity are measured at GDL inlets and outlets with the calibrated instruments. At the dry outlet, a condenser permits us to reduce and to control the water content; thus, the airflow and the temperature are measured using a calibrated flow meter assuming saturation.

2.2. Results reliability evaluation

Table 1 reports the range and the measurement uncertainty of all measured parameters. These quantities were acquired at a frequency of 1 Hz for 1500 s in steady state conditions. The data were processed with a robust method for outlier elimination. The representative values of each parameter were obtained as the average of the first 1000 elements among the remaining ones.

The uncertainty of the experimental setup was evaluated for the measurements of both the water concentrations and the water flow through the GDL. The global uncertainty of the experimental