Journal of Power Sources 285 (2015) 568–579

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

Study of effective transport properties of fresh and aged gas diffusion layers

Magdalena Bosomoiu, Georgios Tsotridis, Tomasz Bednarek*

European Commission, Directorate-General Joint Research Centre, Institute for Energy and Transport, Postbus 2, 1755 ZG Petten, The Netherlands

HIGHLIGHTS

- The brand new, conditioned and aged GDL samples are reconstructed using CT to obtain the effective tensor material parameters.
- The effect of GDL compression by Bi-Polar plates' ribs is considered.
- The water saturation and its impact on GDL transport properties are investigated.

ARTICLE INFO

Article history:
Received 24 September 2014
Received in revised form
11 March 2015
Accepted 21 March 2015
Available online 24 March 2015

Keywords:
Gas diffusion layer degradation
Effective properties
Proton exchange membrane
Fuel cell
Computer tomography

ABSTRACT

Gas diffusion layers (GDLs) play an important role in proton exchange membrane fuel cells (PEMFCs) for the diffusion of reactant and the removal of product water. In the current study fresh and aged GDLs (Sigracet® GDL34BC) were investigated by X-ray computed tomography to obtain a representative 3D image of the real GDL structure. The examined GDL samples are taken from areas located under the flow channel and under the land. Additionally, a brand new Sigracet® GDL34BC was taken as a reference sample in order to find out the impact of fuel cell assembly on GDL. The produced 3D image data were used to calculate effective transport properties such as thermal and electrical conductivity, diffusivity, permeability and capillary pressure curves of the dry and partially saturated GDL. The simulation indicates flooding by product water occurs at contact angles lower than 125° depending on sample porosity. In addition, GDL anisotropy significantly affects the permeability as well as thermal and electrical conductivities. The calculated material bulk properties could be next used as input for CFD modelling of PEM fuel cells where GDL is usually assumed layer-like and homogeneous. Tensor material parameters allow to consider GDL anisotropy and lead to more realistic results.

1. Introduction

In recent years, the use of low temperature proton exchange membrane (PEM) fuel cells has increased considerably for automotive applications, back-up power units and small portable units because of their high efficiency, high power density, absence of emissions, low operating temperature and low noise. The single cell PEM fuel cell consists of one membrane electrode assembly (MEA) located between two flow plates with the role of providing the reactant gas access, controlling the temperature and collecting the current [1].

The Gas Diffusion Layer (GDL) is one of the components of the MEA and is placed at each side of the MEA. GDL is a support for catalyst but this study focuses on its transport properties. GDL plays an important role in gas transport from the flow channels to the catalyst layer, removal of the produced water, electron transport between catalyst layer and bi-polar plate and reaction heat removal. The GDL consists of carbon fibres that are impregnated with polytetrafluoroethylene (PTFE) to ensure that the hydrophobic character of the GDL is maintained.

The models developed for studying fuel cell performance are macroscopic models making simplifications of the real structure of the GDL [2–10]. Lemoine-Nava et al. [11] made an analysis of flooding and drying of the GDL and the catalyst layers by using a 1D transient two-phase flow model. It was shown that severe flooding of the GDL is mainly due to poor removal of liquid water (determined by the material properties) rather than to high water condensation rate, thereby highlighting the necessity to correctly

* Corresponding author.
E-mail address: Tomasz.Bednarek@ec.europa.eu (T. Bednarek).

http://dx.doi.org/10.1016/j.jpowsour.2015.03.132

estimate the material properties. Since water is produced in the cathode catalyst layer, there is a higher susceptibility of cathode side flooding. However, water has a controversial effect: too low content will have an adverse impact on fuel cell operation, causing an increase in ohmic resistance (due to the reduction in protonic conductivity of the membrane), while high water content will obstruct the diffusion path of the gases. In addition, due to the GDL anisotropy, the properties might vary significantly through plane and in-plane. Therefore, when performing computational fluid dynamics (CFD) modelling, the dependence of transport properties on the local saturation levels and on the structure of the material are very important to achieving reliable results.

The water management in PEM fuel cells was investigated in Refs. [12,13]. The morphology model was used to examining the effects of liquid water presence on the transport properties of the carbon paper GDL. The effects of porosity and fibre distribution on the effective thermal conductivity were also studied. Calculations of effective thermal conductivity and diffusion coefficient in case of MPL were the goals of the papers [14–16]. Stochastic models were used to make a 3D reconstruction of the GDL and MPL. The effects of internal structure of the GDL and MPL (Micro Porous Layer) on transport properties (e.g. diffusion coefficient and thermal conductivity) were investigated. The effects of porosity and fibre distribution on the effective thermal conductivity were studied by Zamel et al. [14]. It was found that thermal conductivity increases with decreasing of porosity and is highly dependent on fibre distribution.

Similar approach was shown by Becker et al. in Ref. [17] in case on MPL. The diffusivity results for reconstructed GDL material with various MPL thicknesses and without MPL were considered. A method was presented which allows to compute binary diffusivity for a range of Knudsen numbers.

Several of the above mentioned papers ([12–17]) deal with investigation of the effective properties for numerically generated GDL or MPL structures.

In order to better estimate the effective transport properties as a non-scalar properties, the 3D geometry of a real GDL should be used in simulations instead of artificially reconstructed structures. Such non-scalar quantities may subsequently be used in CFD calculations where the GDL is simply treated as a homogeneous layer of constant porosity. This would allow to perform 3D fuel cell simulations taking into account the anisotropy of fuel cell parts, especially GDL.

The main objective of this study is to show the influence of fuel cell assembly and ageing processes on the effective GDL transport parameters, such as: electrical and thermal conductivities, diffusivity, capillary pressure as well as gas permeability. Several liquid water saturation levels were also taken into consideration. The X-ray computed tomography technique (CT) was used to determine the 3D anisotropic structure of different GDL samples such as fresh and aged; located under the flow channel and under the land, at a resolution of 1.5 μm. The results of the two aged materials – taken from under the channel and under the land regions – were compared with the fresh ones. The effect of liquid water is studied by calculating the saturation dependence transport properties.

2. Preparation of samples

In this study three types of materials were used, namely: i) One type called "fresh". This material was extracted from the MEA that was installed in a fuel cell setup but never operated. The purpose of using this material was to study the effect of compression to the GDL. ii) The second type called "aged". This GDL was extracted from an MEA that was operated for 1000 h at varying loads between 20% and 100% of the maximum power. iii) The third type called "brand new". In contrast to the other materials, this GDL was not extracted from MEA. The brand new Sigracet® GDL34BC sample is taken from GDL sheet provided by SGL GROUP company [18].

The fresh and aged GDLs, as part of the MEAs, were installed in a fuel cell setup. The fuel cell was assembled by applying a torque of 10 Nm on each of the bolts of the fuel cell setup according to manufacturer requirements. During the assembly process some areas of the GDL are located under the channel and they are called thereafter "under channel", other ones are compressed by the Bi-Polar plates ribs and are called "under land", see Fig. 1.

A sample with a width of 3 mm and a length of 10 mm were cut from each MEAs, fresh as well as aged. In order to study the compression of the GDL effect on the bulk properties, the MEA samples were cut along the lines marked by the Bi-Polar plates during fuel cell assembly, which could be easily observed on the GDL surface. Such method of sample preparation allows the examination of the GDL located under the channel as well as under the land.

These MEA samples are investigated using an X-ray computer tomograph Phoenix Nanotom S system that includes a 180 kV nanofocus X-ray tube and a 2D X-ray detector with 120×120 mm active area (2300×2300 pixels). Since the diameter of MPL pores are of the order of nm [15] the resolution of the CT scan was not sufficient to reliably reproduce the MPL, therefore, the MPL was excluded from considerations in this study.

Excision of the MPL from Sigracet® GDL34BC implies that properties of Sigracet® GDL34BA could be taken into account. The only difference between Sigracet® GDL34BC and Sigracet® GDL34BA is the MPL. The"BC" marking means GDL with MPL whereas "BA" corresponds to GDL without MPL. The carbon paper layer is the same in both, "BC" and "BA" GDLs, see Fig. 1 [18].

The numerical images used in the simulations are taken from the cathode side of the MEAs by taking into account the under the channel and under the land areas. In order to avoid influence of MPL, possible membrane impregnation and surface effects (for example loose fibres) the 3D GDL models are taken from the middle of the scanned cathode GDL, see Fig. 1. Due to above mentioned reasons the thickness of the GDL numerical model is limited to 150 μm as compared to the thickness of brand new Sigracet® GDL34BC is 315 μm. The location of the investigated samples is shown in Fig. 1. The total size of each samples are $150 \times 600 \times 1200$ μm.

In total, there are four examined GDL samples taken from cathode side of fresh and aged MEAs, namely: fresh under channel, fresh under land, aged under channel and aged under land. Additionally, the brand new Sigracet® GDL34BC sample was taken as a

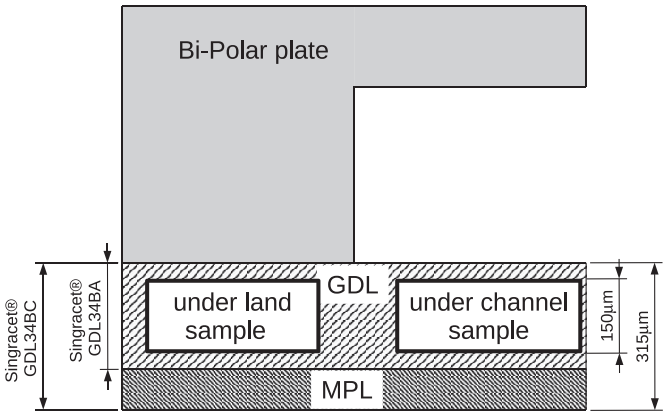

Fig. 1. The location of the investigated samples in the fuel cell setup.