



ELSEVIER

Available online at www.sciencedirect.com

ScienceDirect

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

Water distribution in high temperature polymer electrolyte fuel cells

Uwe Reimer^{a,*}, Jannik Ehlert^a, Holger Janßen^a, Werner Lehnert^{a,b}

^a Forschungszentrum Jülich GmbH, Institute of Energy and Climate Research, Electrochemical Process Engineering, 52425 Jülich, Germany

^b Modeling in Electrochemical Process Engineering, RWTH Aachen University, Germany

ARTICLE INFO

Article history:

Received 18 August 2015

Received in revised form

12 November 2015

Accepted 19 November 2015

Available online xxx

Keywords:

High temperature polymer

electrolyte fuel cell

Water transport

Gas crossover

ABSTRACT

In this work a high temperature polymer electrolyte fuel cell based on a phosphoric acid doped polybenzimidazole membrane is operated at 160 °C on dry air and dry hydrogen. The anodic stoichiometry was varied to resemble typical operation with pure hydrogen or reformat gas. At the outlet of the fuel cell liquid water was collected by condensers. The resulting amounts of liquid water from cathode and anode were computationally analyzed. The results yielded an effective diffusion coefficient of water vapor of $D \approx 2.7 \cdot 10^{-7} \text{ m}^2 \text{ s}^{-1}$, which is the average value for the cell based on the water partial pressures at the outlets. From the calculated water partial pressures at the anode and cathode it can be concluded that the concentration of phosphoric acid differs significantly within catalyst layer of the anode and cathode. Finally, the crossover of hydrogen and oxygen was shown to depend on the swelling of the membrane.

Copyright © 2015, Hydrogen Energy Publications, LLC. Published by Elsevier Ltd. All rights reserved.

Introduction

Polymer electrolyte fuel cells (PEFC) are energy converters that achieve their greatest efficiency if hydrogen is used as fuel. During the conversion process water is produced in the cathode electrode layer [1]. For Nafion type PEFCs, which are operated up to 80 °C, the overall distribution of water is a highly complex process. This has been summarized by several review papers describing the overall effects [2–5] as well as modeling aspects [6–8] and liquid water visualization [9]. Water leaves the fuel cell both in a liquid and vaporous state. Therefore, evaporation and condensation effects are important in the fuel channels, gas diffusion layers (GDL) and electrode layers. The application of Nafion membranes also

requires the humidification of the gases in order to ensure good protonic conductivity at the inlet region. Furthermore, liquid water is transported across the membrane by means of the following processes [3]:

- *electro-osmotic drag* due to the hydration shell of hydronium ions,
- *back diffusion* due to concentration differences between the anode and cathode catalyst layers,
- *capillary effects* in the gas diffusion and catalyst layers and
- *hydraulic permeation* if a pressure gradient exists between the anode and cathode sides.

The convective mass transfer in the reactant flow field also affects water distribution between anode and cathode sides.

* Corresponding author.

E-mail address: u.reimer@fz-juelich.de (U. Reimer).

<http://dx.doi.org/10.1016/j.ijhydene.2015.11.106>

0360-3199/Copyright © 2015, Hydrogen Energy Publications, LLC. Published by Elsevier Ltd. All rights reserved.

Depending on the specific design and operation conditions water can be more shifted either to the cathode or anode side. Moreover, the ratio of liquid water against water vapor at the outlets can be influenced.

In high temperature polymer electrolyte fuel cells (HT-PEFC) water management is simpler, since only water vapor is present in the gas channels and GDL. This work considers an HT-PEFC based on phosphoric acid doped polybenzimidazole membranes. This fuel cell type is usually operated at 150 °C to 180 °C [10–12]. The electrolyte is phosphoric acid, which also contains a certain quantity of water. The HT-PEFC can be operated with both dry or humidified gases. It has been reported that the humidification of anode gases leads to a small increase in fuel cell performance [13–15], especially if carbon monoxide is present [15]. Conversely, a small decrease of performance was observed under certain conditions [16]. The simultaneous humidification of anode and cathode seems to produce no overall effect [17]. Depending on the operating conditions, it is possible to observe active water transport from either cathode to anode or vice versa [16,18].

The effects described above are strongly related to the properties of phosphoric acid as an electrolyte. Orthophosphoric acid is commercially available at concentrations of up to 85 wt%. At this concentration it contains a molar ratio of phosphoric acid/water equal to 1/1. With increasing temperature the acid (actually the acid doped PBI membrane) releases water in two major steps [19]. At temperatures of up to 100 °C water evaporates, leaving a higher concentrated acid solution behind. At temperatures of around 120 °C to 130 °C phosphoric acid starts to form dimers (so called pyrophosphoric acid) as a first condensation product [19,13,20]. With further increasing temperature the condensation process results in a complex mixture of acid species [21,22].

The increased temperature results in two opposing trends. Firstly, membrane conductivity increases with temperature due to increased ionic mobility [23]. Secondly, membrane resistance increases with decreasing water content [16,17]. Therefore, the overall water balance of the fuel cell has some influence on its overall performance. On the other hand, this influence may sometimes be small, as under normal operating conditions excess water is produced at the cathode and readily distributed through the thin membrane. An additional degree of humidification can also lead to a significant volumetric increase of acid in the catalyst layers, thus increasing mass transfer resistance, as described by Chen and Lai [17] and also reported by Galbiati et al. [16]. Furthermore, humidification leads to dilution effects on the reacting gases, which become important for operation in dead end mode [24,25].

In contrast to PEFCs, there seems to be no electro-osmotic drag of water from anode to cathode [26]. Furthermore, overall proton conductivity seems to originate almost entirely from proton transfer via the shift of hydrogen bonds (the so called Grotthuss mechanism) [27–29,19,23].

In the present work, the distribution of water vapor at the outlets of the cathode and anode is analyzed. As reactants serve dry air and dry hydrogen. Volumetric flow rates are chosen to cover the range of typical hydrogen/air and reformate/air operation. The goal is to clarify the main driving force of water distribution when phosphoric acid is used as an electrolyte.

Experimental

For the experiments a commercial membrane electrode assembly was used (CELTEC P1000, BASF 2011). The graphite composite type flow field consisted of a 3-fold serpentine with the anode and cathode in a coflow configuration. The active cell area was $A = 45 \text{ cm}^2$. The cell was operated at $T = 160 \text{ °C}$ and ambient pressure. Temperature was controlled by two electric heaters inside the metallic end plates. A sketch of the overall setup is shown in Fig. 1. The supplied gases were dry hydrogen and dry air with a dewpoint of -61 °C . Mass flow controllers of the type F-201 (Bronkhorst [30]) with $\Delta m < 1.0 \%$ were used to supply air and hydrogen. The temperature of the coolant of the condensers was set at 7 °C . Liquid water was collected in closed bottles during at least 70 h of continuous operation for each operation point and subsequently weighted by a laboratory scale at $\Delta m = \pm 0.01 \text{ g}$.

In order to cover the normal operation range, measurements were performed under constant load at $j = 0.2 \text{ A cm}^{-2}$, 0.4 A cm^{-2} and 0.6 A cm^{-2} . The cathode stoichiometry was fixed at a ratio of $\lambda_c = 2$. The anode stoichiometry ratio was $\lambda_a = 2, 4$ and 6 in order to mimic typical flow rates that correspond to operation with reformat gas.

Results and discussion

Underlying assumptions

Interpretation of the experiments is based on the following general assumptions. Water is produced inside the cathode catalyst layer of the HT-PEFC. It is assumed that the water is produced within a layer of phosphoric acid that covers the catalyst, i.e. water is virtually in a liquid state. The concentration of phosphoric acid decreases as a function of water production. Water can leave the fuel cell by evaporation through the cathode GDL and gas channel (see Fig. 2). On the other hand, the difference in phosphoric acid concentration gives rise to a diffusion flux of water from the cathode catalyst layer to anode catalyst layer. At the anode, water leaves the fuel cell through evaporation. The total water balance is determined by local current density (water production), local flow rates within the catalyst layers (evaporation rate) and the concentration gradient of phosphoric acid in the membrane electrode assembly (diffusion flux).

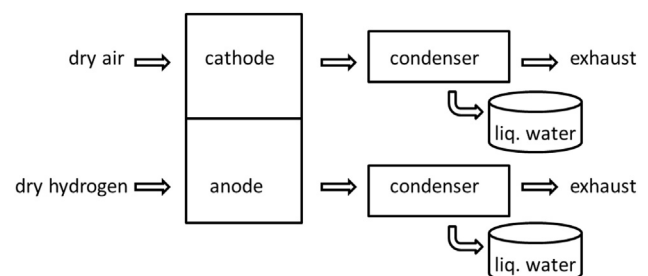


Fig. 1 – Sketch of the experimental setup.

Download English Version:

<https://daneshyari.com/en/article/7712423>

Download Persian Version:

<https://daneshyari.com/article/7712423>

[Daneshyari.com](https://daneshyari.com)