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Experimental study of water transport in a polybenzimidazole-based high temperature PEMFC

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ABSTRACT

The present work reports a systematic experimental analysis on water transport in a phosphoric acid doped polybenzimidazole-based high temperature PEM fuel cell. Two sets of polarization curves are run with dry and alternatively humidified reactants, covering a wide range of fuel cell operating temperatures and stoichiometries. With dry feed streams, up to 18% of water produced by electrochemical reaction is found on anode side proving the presence of water transport from cathode electrode. Under the investigated conditions, water transport across the membrane is independent of fuel cell temperature but strongly dependent on reactants stoichiometry and humidification. Such parameters can even determine a change in water transport direction. Humidification causes a limited drop in membrane proton resistivity (around 6 mΩ cm^2); conversely a slight decrease in fuel cell performances (−5 to −20 mV) is measured.

1. Introduction

In the last two decades research devoted increasing attention to high temperature polymer electrolyte membrane fuel cells (HT-PEMFC) as a possible alternative to conventional PFSA membrane based low temperature fuel cells [1–5]. Higher operating temperature, ranging from 120 to 180 °C, actually allows higher tolerance to fuel impurities and easier fuel cell cooling thus improving catalysts resistance to pollutants and heat recovery [3,5–7]. Therefore this technology is considered very promising for natural gas fed CHP systems with an integrated fuel processor [6,8–10]. Furthermore the introduction of acid doped polymers as electrolytes reduces the dependence of proton conductivity on polymer hydration resulting in unnecessary reactants humidification. Such characteristics could simplify fuel cell based energy systems with a possible positive consequence on their cost reduction and diffusion. Up to now a large number of studies has been carried out on HT-PEMFCs mainly analyzing performances [2,7,11,12], degradation [9,13–19], membrane properties [1,3,20–24], acid distribution [25–27] and internal losses [28,29]. Nevertheless, several aspects remain unclear and need to be further investigated in order to achieve deeper knowledge. Particularly important is the effect of water transport on HT-PEMFC behavior. Actually, even though HT-PEMFCs can operate in dry conditions, it does not occur in real applications where a high water content is always present in syngas produced by fuel processors. The eventual necessity of fuel drying would imply an expensive water removal system, thus HT-PEFCs tolerance to humidified conditions needs further investigation to clarify some concurrent effects. Water is reported to improve proton conductivity both in the membrane and in the electrodes due to its effect on acid equilibrium [3,26,30–33]. Furthermore water on anode side can contribute

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to water-gas shift reaction to oxidize CO when reformed gas is used [34]. On the other side, supplying water through the humidification of feed streams decreases reactants partial pressure [35,11] and can increase acid leaching from electrodes with negative effects on performances due to a reduction in the three phase zone [33]. Therefore quantifying water transport within the MEA in dry and humidified conditions is of relevant interest. Reactants humidification is a useful instrument to enhance the effects of water presence even if running an HT-PEMFC with humidified reactants appears questionable. In the present work an experimental analysis of internal water transport in a phosphoric acid doped polybenzimidazole (PBI) based HT-PEMFC is presented. Effect of cell temperature, stoichiometries and current density is analyzed when dry and humidified reactants are fed. Measurements uncertainty, repeatability and mass balance are analyzed, according to [36], to assure for reliable results interpretation.

2. Experimental

2.1. Methodology

A single PBI based HT-PEMFC is operated with pure hydrogen and air in co-flow configuration. After an activation period of 70 h at reference conditions ($T_{cell} = 160$ °C, $i = 0.2$ A cm^{-2}, $\lambda_{H2} = 1.2$, $\lambda_{air} = 2$) as recommended by the manufacturer [37], two consecutive sets of polarization curves are performed during which an extensive range of cell temperatures and reactants stoichiometries is covered. During the first set of polarization curves (Table 1) both reactants are fed as dry, while during the second set (Table 2) humidification of anode or cathode stream is alternatively performed. Polarization curves are run imposing fuel cell current while reactants flow rates are automatically regulated in order to keep stoichiometry constant. All curves are made up by nine current density values ranging from 0.2 to 0.9 A cm^{-2} with constant step; during each test OCV is kept for 100 s and voltage is checked. Current density values lower than 0.2 A cm^{-2} are not considered due to poor accuracy in analyzing mass balance. Each current density value is kept for 1200 s to make sure that cell's internal transport phenomena reach steady state. Data are recorded at 1 Hz frequency and are processed with a robust method for autliers elimination. The method eliminates values not included in the interval: median ±3 times standard deviation, estimated through median absolute deviation (MAD). Representative values for every working point are obtained as average of 600 elements among the remaining ones. In both sets every polarization curve is randomly repeated for three times in three different days to assure for reproducibility, considering measurements uncertainty (Table 3). The Representative value for each measured parameter is obtained as average of the three measures.

Table 1 – Investigated conditions with dry reactants.

T cell [°C]	λ_{H2}	λ_{AIR}
140	1.3	2–4
160	1.2–1.3	1.5–2–4
160	1.5–1.75	2
180	1.3	2–4

Table 2 – Investigated conditions with humidified reactants.

T cell [°C]	λ_{H2}	λ_{AIR}	Humidified side	Saturation Temperature[°C]
160	1.2	2	A	30–40–50
160	1.2	2	C	30–40–50
160	1.2	4	A	30–40
160	1.2	4	C	30–40
160	1.5	2	A	30–40–50
160	1.5	2	C	30–40–50

2.2. Experimental setup

2.2.1. MEA

The object of this investigation is a commercial HT-PEMFC (Celtec-P2100 by BASF Fuel Cells) based on a phosphoric acid doped PBI membrane. This fuel cell has an active area of 20 cm^2 and consists in a package of MEA and gas diffusion layers provided with PTFE gaskets (thickness: 345 μm anode, 370 μm cathode). Electrode catalyst is Platinum on anode side (loading: 1 mg cm^{-2}) and Platinum alloy on cathode side (loading: 0.7 mg cm^{-2}). Membrane doping level is reported to be up to 70 phosphoric acid molecules per PBI repeating unit [18].

2.2.2. Fuel cell assembly

The MEA is placed between two graphite distributors with carved serpentine flow fields (single serpentine on anode side and triple serpentine on cathode side, both with the same square section: depth and width 0.8 mm, length 700 mm; ribs width: 0.8 mm). Graphite distributors are held together between two stainless steel plates connected with 8 screws closed with a controlled torque of 12(±0.5) Nm. Temperature of plates is measured by a calibrated thermocouple (uncertainty: 1 °C, distance from active area: 20 mm) and is controlled by a temperature controller connected with two cartridge heaters located inside of the steel plates as well. Since heat capacity of the plates is much higher than that of graphite distributors and MEA, high temperature stability is attained. Furthermore, due to the high thermal conductivity of graphite distributors and to the MEA small thickness, temperature uniformity in the entire fuel cell assembly is assumed.

Table 3 – Uncertainty of measured parameters.

Parameter	Estimated uncertainty
Fuel Cell Voltage	7 mV
Water Concentration	5%
Cathode Exhaust Water Flow	4.7% + 2.5 10^{-8} mol s^{-1} cm^{-2}
Anode Exhaust Water Flow	7.1% + 7 10^{-9} mol s^{-1} cm^{-2}