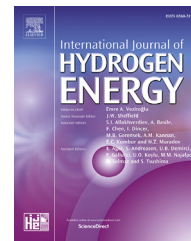


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New load cycling strategy for enhanced durability of high temperature proton exchange membrane fuel cell

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ABSTRACT

The objective of this paper is to develop a new operational strategy to increase the lifetime of a high temperature proton exchange membrane (HT-PEMFCs) fuel cell system by using load cycling patterns to reduce the phosphoric acid loss from the fuel cell. Four single cells were operated under different current cycling profile, while one cell was operated at constant current density for comparison. Polarization curves and electrochemical impedance spectroscopy measurements were recorded during the course of the tests and analysed. Two different current densities, 0.2 Acm^{-2} for the lower end and 0.8 Acm^{-2} for the higher end, were selected for the load cycling operation. The relaxation time, which is the period of time spent at low current density operation, is varied to understand how the performance over prolonged period behaves. The duration of the high current density operation is selected based on the relaxation time in order to have the same average current density of (0.55 Acm^{-2}) for all the cells. Cell 5, with a relaxation time of 2 min performs best and shows lower degradation rate of $36 \mu\text{Vh}^{-1}$ compared to other load cycling cells with smaller relaxation times. The cell operated at constant current density shows a degradation rate of $57 \mu\text{Vh}^{-1}$, which is 1.5 times higher than the degradation rate of cell 5.

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Introduction

High temperature polymer electrolyte membrane fuel cells (HT-PEMFCs) are promising combined heat and power (CHP) sources with a higher combined efficiency compared to conventional low temperature PEM fuel cells. The advantages of HT-PEMFCs are various in terms of higher tolerance to

poisonous gases like CO and H₂S due to decreased adsorption and faster reaction kinetics [1], no or less complex humidification system required as the proton conductor in these type of fuel cells are phosphoric acid (PA) and not water. They also generate useful heat, which can be utilized and the combined CHP efficiency is quite high (>75% upto 85% as reported in these literatures) [2,3]. However, there are also some hurdles, with durability and reliability being the major ones, due to

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different degradation mechanisms, including loss of electrolyte. The phosphoric acid (PA) in HT-PEMFCs, which acts as an electrolyte is quite mobile. This leads to acid loss during different operating conditions, and therefore, the proton conductivity of the membrane is affected negatively.

Lang et al. [4], carried out long and short-term degradation tests to quantify the effect of acid loss on the performance of an HT-PEMFC. The degradation was higher for short water stress test, which involved higher humidification on the anode and cathode. The test results in terms of impedance and polarization measurements confirmed that the cell degradation was dominated by the acid leaching from the membrane. Rastedt et al. [5] carried out different operational strategies to understand the effects on PA loss. The different operational modes included start/stop, different fuel compositions and long time durability tests. The results indicate minimal effect on PA loss with varying operational strategies. Wannek et al. [6] carried out experiments to understand the influence of acid injecting methodology. The analysis showed no or minimal differences in performance with pre-doped or post-doped MEAs. It was reported that the acid redistribution to reach an equilibrium took several hours. However, the acid distributed on the membrane within a few hours is enough to draw reasonable performance.

Chevalier et al. [7] developed a pore network model to understand the role of micro porous layer (MPL) in the redistribution of PA in the gas diffusion electrode (GDE). The model predicts that the presence of MPL mitigates partially the acid leaching from the catalyst layer (CL) to the flow-field. Galbiati et al. [8] investigated the activation period with different procedure to understand the break-in. An MEA exposed to atmosphere before activation procedure showed a loss but the voltage development during activation was recorded same.

Eberhardt et al. [9] investigated the redistribution of PA in an HT-PEMFC using X-ray tomographic microscopy imaging. The most influencing factor for acid movement from the cathode to the anode was acid doping level and the current density. At higher doping levels and high current density ($>0.4 \text{ Acm}^{-2}$), the migration rate increased significantly. In another work by the same group [10], the effect of different operating parameters on the PA loss by evaporation was evaluated by condensation at outlet and accelerated stress test. The parameters considered were current density, gas flow rate and cell temperature. Based on the experiments, a lifetime prediction was calculated as a function of current density and temperature [10]. The influence of current density, different materials and varying PA content was examined to determine the effect of PA migration. The increase in current density above 0.4 Acm^{-2} showed an increase in the migration of acid from the anode to the cathode under the influence of higher capillary pressure from the GDE pores. Also, a higher PA loading leads to higher acid migration, with similar behaviour for the different materials [11].

Eberhardt et al. [12], investigated synchrotron-based X-ray tomographic to determine the local distribution of PA in HT-PEMFC. The method was developed at room temperature, which they proposed could be further applied in-operando cells with slight modifications to understand the PA migration and redistribution. The same authors [13] investigated the dynamic operation PA redistribution under two different load

profiles. The acid migration from the cathode to anode under the influence of current was shown using imaging. The acid migration towards anode GDE and flow-field was reported to be faster than the back diffusion of PA under low current density (0.2 Acm^{-2}). The process of acid migration is shown to be not completely reversible, some of the acid gets stuck to the flow field and is removed by the gas.

The focus to date has been on understanding the PA migration and redistribution. Only limited number of mitigation strategies has been proposed in the literature. The proposed strategies mostly focus on modified GDE/MPL structures [14–16].

In this paper, we propose a new operation based strategy to deal with the acid movement towards the anode and thereby decreasing the degradation due to acid loss. The efficiency calculated from the operation is improved marginally. Current cycling tests between a high current density of 0.8 Acm^{-2} and a low current density of 0.2 Acm^{-2} were performed in order to understand how the electrolyte loss could be avoided. The rest time, also described as relaxation time in this work is defined as the period of time the cell is operated at a low current density of 0.2 Acm^{-2} in order to relieve the cell of the high current density stresses.

The focus was to determine the time constants for the flooding and de-flooding of the anode GDE by the migration of acid from the cathode to the anode. The current load strategy is modified to prevent the acid moving towards anode from getting detached and thereby keeping the acid as much as possible within the cell for a longer lifetime. An increase in lifetime is predicted based on the impedance and the voltage inventories.

Experimental setup and procedure

This section presents the experimental setup and the procedure followed for the tests. To verify the effect of load cycling on HT-PEM fuel cells, the method is tested on five membrane electrode assemblies (MEAs), where only the duty cycle of the load cycling has been changed. The tests are carried out to evaluate different operational methods to improve the efficiency and lifetime of an HTPEM fuel cell system. The focus is to retain PA in the membrane for a longer period by reducing the PA losses, while also running at higher current density. During the experiments, the average current density is kept constant for all 5 MEAs. The MEAs tested in this work are phosphoric acid-doped PBI-based with platinum as catalyst manufactured by Danish Power Systems®.

Test setup

The MEAs are tested in an in-house built five single cell test stand, shown in Fig. 1. The test stand has the ability to record impedance spectroscopy, I–V curves and perform load cycling with varying duty cycles. The assemblies are heated to $160 \text{ }^\circ\text{C}$ by electric heaters, which are controlled by the fuel cell control system. Two different computer systems handle the data acquisition: an NI-9024 cRIO which controls the fuel cell sequence, temperature, gas mass flow etc. and an NI real time system with a PCIe-6259 installed, controls the electrochemical

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