H_2 CROSSOVER (H2X)

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Membranes are responsible for conducting protons and separating gasses between the anode and cathode electrodes of a fuel cell stack. As the fuel cell ages, pinholes form in the membrane, which incurs detrimental gas crossover between the electrodes; mainly hydrogen crossover from anode to cathode. Accurate measurement of the leak rate in each individual cell is an essential prerequisite to design and integrate fuel cell components and systems for improved performance and longevity. The Greenlight H_2 Crossover (H2X) module is an add-on to Greenlight's Emerald control and automation software, and is capable of *in-situ* measurement of hydrogen transfer leaks in individual cells of a fuel cell stack.

Background

There are two transport processes that contribute to gas crossover through membranes: permeation and convection. Permeation is driven by a concentration gradient of gasses across the membrane and is proportional to the diffusion coefficient of the gasses in the membrane. Convective transport occurs only when pinholes are formed in the membrane, and its rate is proportional to the pressure gradient across the pinhole as well as the viscosity of the gas. Permeation is the primary mechanism for gas crossover in membranes without pinholes, whereas the convective mechanism becomes dominant as pinholes develop.

The rate and direction of the convective crossover mechanism can be controlled by the overpressure between anode and cathode electrodes. Cathode overpressure would result in oxygen crossover to the anode, which would combine with hydrogen and could result in anode starvation. Anode overpressure, however, would result in hydrogen crossover to the cathode, which would combine with oxygen and could result in cathode starvation. Furthermore, hydrogen starvation can cause cell reversal and irreversible damages to the cell, whereas oxygen starvation would result in mostly reversible losses. Therefore, fuel cell stacks are operated with a small anode overpressure, in order to protect against hydrogen starvation once pinholes are formed. As a result, crossover leaks are considered hydrogen crossover.

There are a number of techniques that can be used to measure the crossover leak rates in fuel cells such as direct measurement, electrochemical leak detection, linear sweep voltammetry, and H2 Crossover (H2X). As discussed in Greenlight's Q_{leak} white paper, H2X is the only technique that can quantify the hydrogen crossover leak rates of individual cells of a fuel cell stack. Therefore, this is the methodology employed by Greenlight's H2X software module.

Methodology

In order to estimate the leak rates of hydrogen in individual cells of a PEM fuel cell stack, the anode and cathode are supplied with hydrogen and nitrogen, while an overpressure in maintained between the two electrodes.

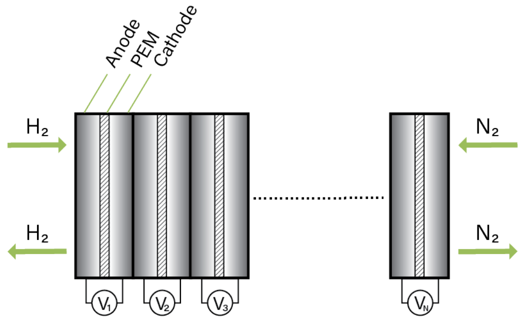

Fig. 1. Schematic diagram of the H2X diagnostic methodology

Under these conditions, hydrogen will crossover from anode to cathode, resulting in a hydrogen partial pressure on the cathode side. The ratio of the anode and cathode hydrogen partial pressures results in a cell voltage that can be obtained using Nernst equation. By accounting for the partial pressure of water in the electrodes, and solving for the hydrogen crossover flow, Q_{leak}, gives [1]:

$$Q_{leak} = \frac{Q_{N_2}}{\frac{P_c - P_{w,c}}{P_a - P_{w,a}} e^{\frac{2EF}{RT}} - 1} \quad , (1)$$

where Q_{N2} is the nitrogen flow, P_c and P_a are the cathode and anode pressures, $P_{w,c}$ and $P_{w,a}$ are the partial pressure of water in the cathode and anode, E is the cell potential, R is the gas constant, and T is the cell temperature.

Equation 1 can be used to estimate the rate of hydrogen crossover flow in individual cells of a PEM fuel cell stack. This rate is a sum of both permeation and convection components. When there is no pinhole in the membrane, this rate would represent hydrogen permeation through the membrane. For large pinholes, permeation can be neglected when compared to convection, and the rate represents hydrogen crossover leak through pinholes. Note that conducting two crossover leak measurements with different overpressures, induced by changing the cathode pressure, can separate the two components. In both cases, as the anode pressure remains constant, the diffusion rate would have negligible change, whereas the convection component would change proportional to the change in overpressure, based on which the permeation and convection processes can be separated. Greenlight's H2X module is equipped with an algorithm that conducts this test automatically, and returns separate values for contributions due to permeation and convection.

As hydrogen crossover flow is proportional to the overpressure, it is important to maintain a fixed overpressure along the cell. This can be achieved by controlling the anode pressure and flow rate to match the cathode pressure drops from input to output. Furthermore, hydrogen crossover needs to be negligible compared to the hydrogen flow rate in the anode, so that there is insignificant disturbance in the pressure drop along the cell due to the crossover leak. Greenlight's H2X module uses an algorithm to ensure these conditions are satisfied.

The H2X module is specifically useful for Accelerated Stress Testing of fuel cell membranes, as they require periodic characterization of hydrogen crossover leak. Furthermore, it can be used to characterize hydrogen permeation through the membrane. The limitation of the technique is that it underestimates the leak rate when pinholes are formed closer to the outlet. The H2X technique is currently being optimized to compensate for this measurement error.

References

[1] *In-situ* diagnostic tools for hydrogen transfer leak characterization in PEM fuel cell stacks Part I: R&D applications, A.M. Niroumand, O. Pooyanfar, N. Macauley, J. DeVaal, F. Golnaraghi, Journal of Power Sources 278 (2015) 652-659.

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