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Open circuit voltage profiling as diagnostic tool during stack lifetime testing

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ABSTRACT

A 10-cell Mk 9 stack was characterized using current/voltage mapping during automotive drive cycle testing. A minimally invasive current mapping technique was used to determine localized polarization curves which together with open circuit voltage (OCV) profile measurements provide useful information about crossover leak formation and location. Through a systematic variation of reactant gas pressures it is further possible to distinguish between electrical shorts, diffusive and convective leaks.

1. Introduction

Advanced diagnostic methods that provide deeper insights into the relationship between cell design, operating conditions and cell performance and degradation are essential in order to meet the cost, durability, power density and freeze-startup targets necessary for PEM fuel cell commercialization. Consequently, AFCC and Ballard have been developing diagnostic test methods related to water management, localized performance measurement as well as the cell voltage loss breakdown (VLB) into kinetic, ohmic and mass transport related components [1,2]. Recently, these methods have been integrated into a fast dynamic test station capable of simulating automotive drive cycles enabling the measurement of current and voltage distributions in real time during dynamically changing operating conditions. By combining current mapping with specific diagnostic protocols, such as for example current–voltage sweeps under air/O_2 operation, local polarizations can be obtained within each cell of a stack. Through a fit of the Srinivasan equation [3] to these local polarizations, the spatially resolved VLB can be determined for each cell. Furthermore, the evolution of the VLB over time together with the change in active Pt-surface area (EPSA) can be used to generate so-called in situ degradation "fingerprints" [4] that are characteristic of a given degradation mode.

This paper illustrates how spatially resolved open circuit voltage (OCV) profiles and current–voltage sweeps can be used to monitor leak formation and growth with spatial resolution and how to determine the type of leak (reactant crossover, electrical short) using a systematic variation of operating pressure. This approach provides valuable failure mode diagnostics and can be used to improve understanding of membrane degradation phenomena during stack lifetime testing.

2. Experimental

A Ballard Mk 9 10-cell stack was equipped with a current mapping setup which consisted of (i) segmented (16-fold) bus plates at both the anode and cathode end and (ii) cell voltage monitoring at eight locations along the cell length, positioned along one of the long sides of the cells (see Fig. 1). This arrangement provided spatial resolution of cell- and in-plane voltages along the fluid flow direction and also enabled the use of standard Membrane Electrode Assemblies (MEA's) and flowfield plates without any modifications (minimally invasive current mapping). The stack was subjected to an automotive drive cycle test that consisted of four 2 h dynamic operation blocks followed by 15 min shutdowns. Operating conditions during drive cycle testing were variable, ranging as follows: RH: 75–90%; inlet temperature: 60 °C; reactant pressures: 15–22 psig; load: 0–1.5 A cm^{-2}; oxidant gas: air; fuel gas: 80–100% H_2 balanced with N_2. The anode and cathode shunts were used to calculate the in(out)going current distribution, the through plane current distribution within each cell can then be calculated using a simple electrical network model once the in-plane resistances R_{jk}^{IP} of the bipolar plates are known. For this, cell voltage measurements at the positions corresponding to the bus plate segments were determined by interpolation using the eight measured values for each cell. EPSA measurements were performed using CO-stripping

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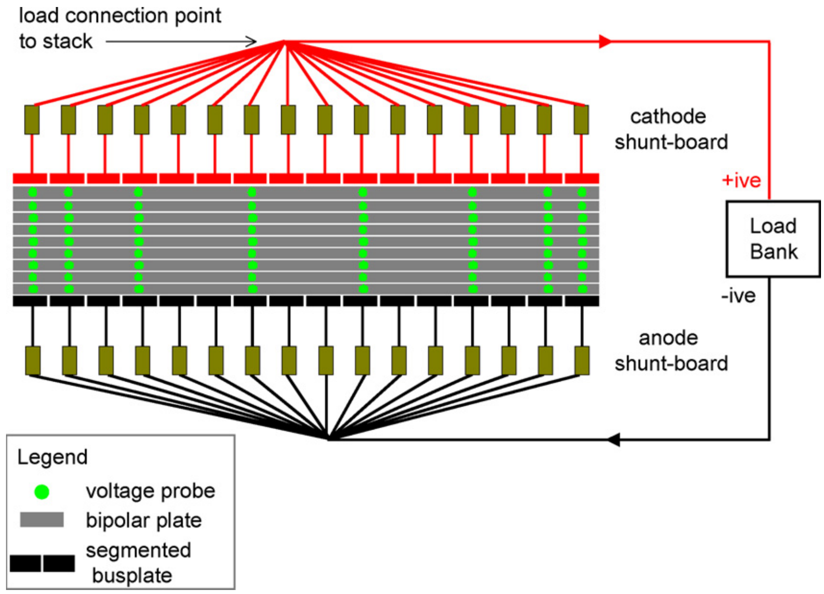

Fig. 1. Schematic diagram of the current mapping setup for the 10-cell Mk 9 stack. Both anode and cathode bus plates are divided into 16 segments or "pucks". In combination with the cell-voltage probing along the cell length this allows current mapping across all cells in a minimally invasive manner.

voltammetry by applying a voltage ramp to the whole stack using a programmable power supply.

In order to determine R_{jk}^{IP}, a special calibration procedure was developed. Ten special "calibration" MEA's were prepared where the membrane was removed selectively only in the areas of segment 1 or 16 in order to create electrical shorts. A stack was prepared with 10 calibration MEA's with shorts alternating in segments 1 and 16 and connected to a power supply to force a current in a zig-zag pattern through each of the bipolar plates. The stack temperature was kept at 60 °C and current–voltage characteristics were obtained by varying the power supply voltage. The observed current–voltage characteristics were linear allowing for the experimental determination of all 176 R_{jk}^{IP} under operating conditions representative of drive cycle operation.

3. Results and discussion

A 10-cell stack was subjected to a dynamic automotive drive cycle and performance diagnostics was performed at various points during duration of the test. The performance diagnostics consisted of cell voltage (I-V polarization) scan, EPSA determination and OCV measurement. Fig. 2 shows the evolution of the average cell voltages at 1.2 A cm^{-2} for each cell during the test. Based on the cell performance in Fig. 2a three distinct phases can be distinguished

1. Run-in or conditioning phase (<15% test duration)
2. Performance phase with only minimal and constant degradation (15% up to 80% test duration)
3. Final degradation phase characterized significantly increased degradation rates and large cell-to-cell voltage variations (80% to 100% test duration)

Fig. 2b shows that the stack voltage degradation in the performance phase is dominated by cells 9 and 10. The phenomenon that end-cells show much higher degradation rates and therefore dominate stack voltage degradation has been observed before and has been attributed to (i) different cell temperatures, (ii) reactant flow mal-distribution or (iii) a combination of the two. It is interesting to note that the high degrading cells are located at the cathode end of the stack, which suggests that liquid water accumulation on the cathode side may be a contributing factor. Fig. 2c shows that the active Pt- surface area on the cathode decays for all cells, with cell 9 exhibiting the greatest degradation (≈40% EPSA loss after 50% test duration). Consequently, catalyst degradation could be an additional cause for the observed high degradation for this cell.

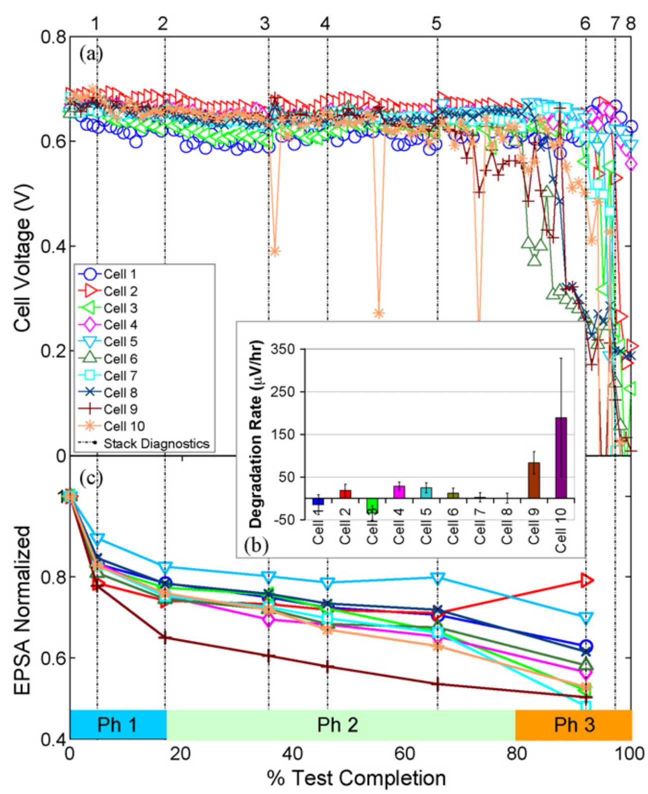

Fig. 2. (a) Lifetime plot of average cell voltage at 1.1 A cm^{-2} for a 10-cell Mk 9 stack during automotive drive cycle testing, vertical dash-dotted lines indicate times of performance diagnostics #1–8. The three performance phases Ph 1–3 are also indicated (see text). Error bars indicate 95% confidence intervals. (b) Average cell voltage degradation rates (negative values indicate performance increase) for cells 1–10 during performance phase (15% up to 80% test duration). (c) Normalized average EPSA loss for cells 1–10 during test duration.