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Short Communication

Tolerance and mitigation strategies of proton exchange membrane fuel cells subject to acetylene contamination



HYDROGEN

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ABSTRACT

Acetylene contamination in proton exchange membrane fuel cells (PEMFCs) significantly depends on cell operating conditions. In this work, acetylene contamination is studied by varying the acetylene concentration, cathode potential and temperature. Hysteresis in the cell performance response is observed during cycling tests with variations in acetylene concentration and cathode potential and is attributed to the potential dependency of acetylene redox reactions. The tolerance of PEMFCs to acetylene is established at approximately 23 ppm for a commercially available catalyst-coated membrane. For a cell poisoned by acetylene at a concentration above the tolerance value, performance losses are eliminated by bringing the ohmic compensated cell voltage into the acetylene oxidation (cell voltage increase) or hydrogenation (cell voltage decrease) regions.

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Introduction

The PEMFC is one of the most promising clean energy technologies and suitable primary power sources for transportation applications [1,2]. Many automobile companies have released their fuel cell vehicles to the automobile market, such as Mirai from Toyota, Clarity from Honda, Tucson FCEV from Hyundai, F-Cell from Mercedes-Benz, and X-Trail FCV from Nissan [3]. All PEMFC stacks in those vehicles are supplied with atmospheric air as the oxidant. Unfortunately, there are over 200 recorded pollutants in air, which may adversely impact the PEMFC performance if the air filter is saturated or damaged. Acetylene, a widely used welding fuel and chemical synthesis reagent, is an airborne pollutant, with a concentration of 5.5 (1 h average) and 3 (24 h average) ppm near production plants [4]. A first study indicated that polluted air containing 300 ppm acetylene causes a cell performance loss exceeding 50% [5]. A tolerance limit needed to design air filters and establish a maintenance schedule, was proposed by extrapolating steady state data [6]. However, this procedure ignored the impact of transient operation, a key automotive application feature.

The acetylene contamination effect significantly depends on operating conditions [7,8]. The cell performance loss increases with increases in acetylene concentration and

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decreases in cell temperature. However, poisoning becomes more severe at intermediate cathode potentials. The contamination mechanism indicates that acetylene adsorbs on the PEMFC electrode and inhibits the ORR in the cathode. When cathode potentials are above 0.65 V vs HRE, adsorbed acetylene is electro-oxidized into carbon dioxide and desorbs easily; when potentials are below 0.3 V vs HRE, adsorbed acetylene is reduced to ethylene, ethane and methane, and the reduction products desorb even more rapidly. However, the electro-oxidation intermediate CO (or COH-type species) formed above 0.5 V vs HRE, and the electro-reduction intermediates vinylidene ($\mu_3 \eta^2$ -C=CH₂) and ethylidyne (μ_3 -C-CH₃) formed below 0.3 V vs HRE. Those intermediates are stable to electro-oxidation, especially vinylidene and ethylidyne. The intermediate potentials caused significant accumulation of adsorbate and redox intermediates on the cathode and significantly hindered the ORR. Those previous results imply that there are concentration and potential barriers: both parameters interact with each other and depend on cell temperature, and they also determine or limit the tolerance of PEMFC to acetylene.

In this paper, the tolerance of the PEMFC to acetylene is determined by gradually cycling the cell conditions; cell performance recovery and mitigation strategies are developed and optimized. Such information is urgently needed for the successful commercialization of fuel cell vehicles subject to transient operation.

Experimental

Experiments were accomplished with a FCATS^{IM} G050 test station (Green Light Power Technologies Inc.), a 50 cm² active area single cell and double/triple serpentine channels for anode/cathode flow fields, respectively [4,5]. Gore PRIMEA M715 catalyst-coated membranes (GCMs) with a Pt loading of 0.4 mg_{Pt} cm⁻² on each side (50% Pt/C, electrochemically active surface area ~70.5 m² g_{Pt}⁻¹) and 25 BC gas diffusion layers (GDLs, SGL Tech.) were used as membrane/electrode assemblies (MEAs) for all acetylene contamination tests. The 100/ 50% relative humidity (RH) H₂/air streams were supplied at a 2/2 stoichiometry. A high H₂ stoichiometry was used to obtain a more stable cell performance, which was deemed appropriate for the study of cathode contaminants. In other words, operating conditions were set to ensure the anode acts as a pseudo-reference electrode. The cell outlet backpressure was 48.3/48.3 kPa for constant temperature tests, and varied from 10/10 to 48.3/48.3 kPa for the temperature ramping tests. These relative pressures correspond to a constant partial pressure for dry reactants of 1 atm.

The cycling experiments were performed by ramping up and down the acetylene concentration and current density and ramping up the temperature within a prescribed duration. A 10 h constant current operation was conducted at 1 A cm⁻² before the cycling tests to confirm stable cell performance. The concentration was ramped from 0 to 500 ppm and then back to 0 ppm at 80 °C and from 0 to 50 ppm and then back to 0 ppm at 45 °C with a current density of 1 A cm⁻². The current density was ramped from 0 (OCV) to 3 A cm⁻² or when the cell voltage reached 0.05 V and then back to OCV with 0, 100, 200, 300, 400 and 500 ppm acetylene in the cathode and at 80 °C. The temperature was increased from 45 to 80 °C after the cell reached a steady poisoning state with 20 or 100 ppm acetylene in the cathode, respectively.

Results and discussion

Fig. 1 a) shows the cell voltage response to the cycling of acetylene concentration from ~0 ppm to 500 ppm at 1 A cm⁻² and 80 °C. Before exposure, the MEA was operated at a constant current density of 1 A cm⁻² to ensure a stable cell performance, approximately 0.68 V, which is only shown as one point at the beginning of the scan (0 ppm acetylene). During the forward cycling (from 0 to 500 ppm), the voltage decreased slowly until the acetylene concentration reached approximately 220 ppm. The voltage loss is approximately proportional to the increase in acetylene concentration. The voltage decrease then began to accelerate until it reached 0.1 V (acetylene above 350 ppm), at which point the voltage decrease slowed and an approximate proportionality with the acetylene concentration was regained. Those results agree with the results obtained at constant acetylene concentration conditions [7,8]. For the backward cycling (from 500 ppm to 0 ppm), the voltage increased slowly until the acetylene concentration decreased to approximately 50 ppm, and the voltage recovery was approximately proportional to the decrease in acetylene concentration. For an acetylene



Fig. 1 — Cell voltage response to a cycled acetylene concentration at a) 80 °C; b) 45 °C.

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