IX.7 Hydrogen Fuel Quality

Tommy Rockward (Primary Contact),
E. Brosha, A. Fernandez, F. Garzon,
R. Mukundan, and C. Padró
Los Alamos National Laboratory (LANL)
P.O. Box 1663
Los Alamos, NM 87545
Phone: (505) 667-9587
E-mail: trock@lanl.gov

DOE Technology Development Manager:
Antonio Ruiz
Phone: (202) 586-0729
E-mail: Antonio.Ruiz@ee.doe.gov

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Objectives

- Test the effects of critical constituents (NH_3, CO, and H_2S) on fuel cell performance and provide data sets to fuel cell modelers to establish predictive mechanistic models.
- Provide guidance to other test facilities to expedite International Organization for Standardization (ISO) TC197 Working Group 12's experimental results.
- Collaborate with international experts to determine levels of constituents for the development of an international standard for H_2 fuel quality.

Technical Barriers

This project addresses the following technical barriers from the Hydrogen Codes and Standards section (3.7) of the Fuel Cell Technologies Program Multi-Year Research, Development and Demonstration Plan:

(I) Conflicts between Domestic and International Standards

(N) Insufficient Technical Data to Revise Standards

Contribution to Achievement of DOE Codes and Standards Milestones

This project will contribute to achievement of the following DOE milestone from the Codes and Standards sub-program section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

- Milestone 26 - Revised (Society of Automotive Engineers/ISO) hydrogen quality guidelines adopted. (4Q, 2010)

Accomplishments

- Developed baseline test procedures for commercial membrane electrode assemblies (MEAs).
- Continued providing data sets to and interacting with fuel cell modelers.
- Performed multiple experiments using both common commercially-available and LANL 'in-house' MEAs.
- Tested critical constituents (NH_3, CO, and H_2S) and presented data sets to the working group and interacted with fuel cell modelers at various meetings.
- Participated in the development of the North American Team's perspective on the levels of the critical constituents in the hydrogen fuel specification.

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Background

LANL scientists have continued their support of the DOE/Energy Efficiency and Renewable Energy Office of Fuel Cell Technologies, in the Safety, Codes and Standards sub-program, through their expertise in the various disciplines such as materials physics and applications, chemistry, and modeling.

There has been an on-going effort to harmonize at the international level, a standard for hydrogen fuel quality, specifically for vehicle applications. This effort is being coordinated by the ISO Technical Committee on Hydrogen (TC197) in Working Group 12 (WG-12). They have maintained a lead role in the collaborative development and implementation of international performance-based codes, standards and regulations based on actual fuel cell performance.

Approach

We attempt to address two important concerns as the standard is being developed and finalized. The first is to recall that the current level of the constituents in the hydrogen fuel specification was based on detection limits. Since the specification origination, analytical techniques have improved considerably, inherently lowering these detectable limits. And, the other concern is that experimental data were collected using different membrane electrode assemblies (MEAs).

This was mainly due to the lack of a commercial supplier to provide platinum loadings desired by the WG-12. We previously developed and performed baseline test protocols for the commonly agreed upon commercial MEAs. Our previous results with the most deleterious critical constituents (NH_3, CO, and H_2S) employed LANL's MEAs and showed unacceptable performance losses. These tests were conducted at the original specification levels. More recently, the WG-12 tentatively agreed to lower both H_2S and CO levels in the fuel specification. Our results shown reflect test with both LANL and the common MEAs at the initial and reduced levels. Table 1 shows the initial and reduced critical constituents taken from the fuel specification.

TABLE 1. Initial and Reduced Critical Constituents taken from the Fuel Specification

Previous Levels	Current levels
NH_3: 0.1 ppm	NH_3: 0.1 ppm
H_2S: 0.004 ppm	H_2S: 0.001 ppm
CO: 0.2 ppm	CO: 0.1 ppm

Results

Test using NH_3, CO, and H_2S as isolated contaminants, in addition to testing combinations of these contaminants were continued. Experiments were conducted using 0.1 mg Pt/cm^2 at the anode and 0.2 and 0.4 mg Pt/cm^2 at the cathode electrodes for LANL and the common MEAs, respectively. We used 50 cm^2 hardware from Fuel Cell Technologies (Albuquerque, NM) with Nafion® (NRE212) as the membrane and our catalyst was 20% Pt/C (ETEK) for the LANL cell and commercial MEAs were provided by W.L. Gore & Associates. In each result shown we operated the cell in constant current mode at 80°C with 83% utilization for the hydrogen and 50% for the air. In Figures 1 and 2, fuel cells with the common MEA were exposed to 4 ppb H_2S and the ISO critical contaminant mixture (NH_3, CO, and H_2S) in the anode feed stream while operating at 100% relative humidity (RH), respectively. After approximately 100 hours of exposure to H_2S, the voltage decay begins and slowly increases over time. This is consistent with an accumulative effect that has been observed with sulfur-platinum interactions. A similar phenomenon occurs in Figure 2, except after 100 hours the decay rate is higher than a fuel cell operating with just a sulfur containing compound. This indicates that the critical contaminants as a mixture are more detrimental to fuel cell performance.

We also probed the impact of RH on fuel cell behavior in the presence of these critical contaminants. Figure 3 illustrates the results of varying the level of humidification. Clearly, the left graph show a slower voltage decay rate and the extent of performance losses is much lower. This implies that the local water will definitely play a major role in the fuel cell behavior operating in the presence of the more soluble impurities.

As mentioned earlier, the initial specification was reduced and tests at those levels (see Table 1) are currently underway. Our preliminary results are shown in Figure 4. Parallel testing with both LANL and the common MEA are being investigated. Results shown are not conclusive, however, they show promise.

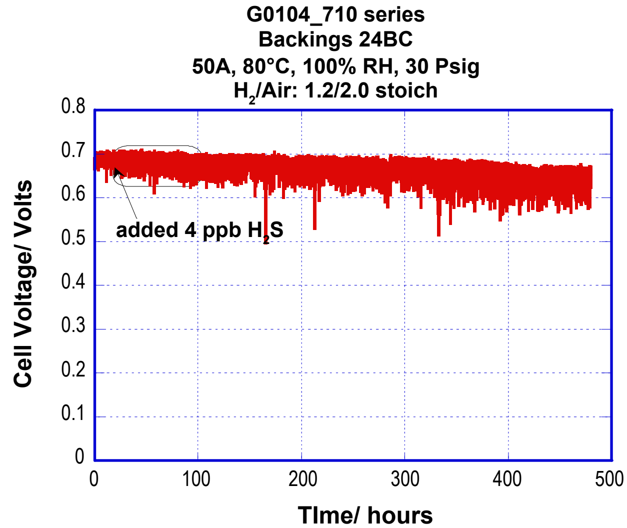

FIGURE 1. Common MEA tested with 4 ppb H_2S at 80°C and 100% RH in a fuel cell operating at 1 A/cm^2.

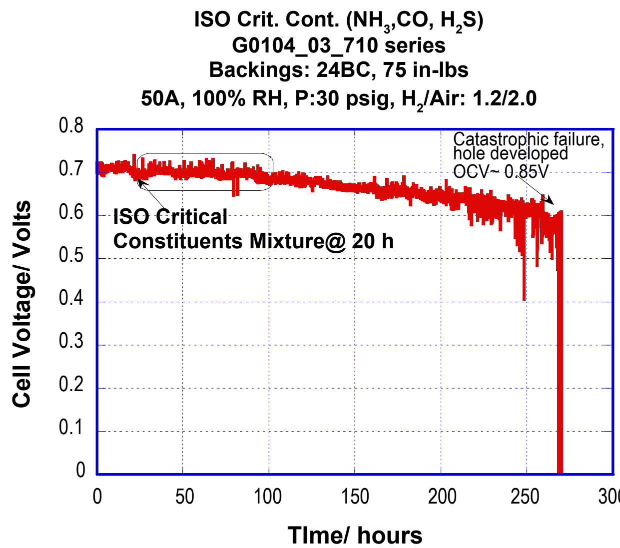

FIGURE 2. Common MEA tested with 4 ppb H_2S, 100 ppb NH_3, and 200 ppb CO at 80°C and 100% RH in a fuel cell operating at 1 A/cm^2.

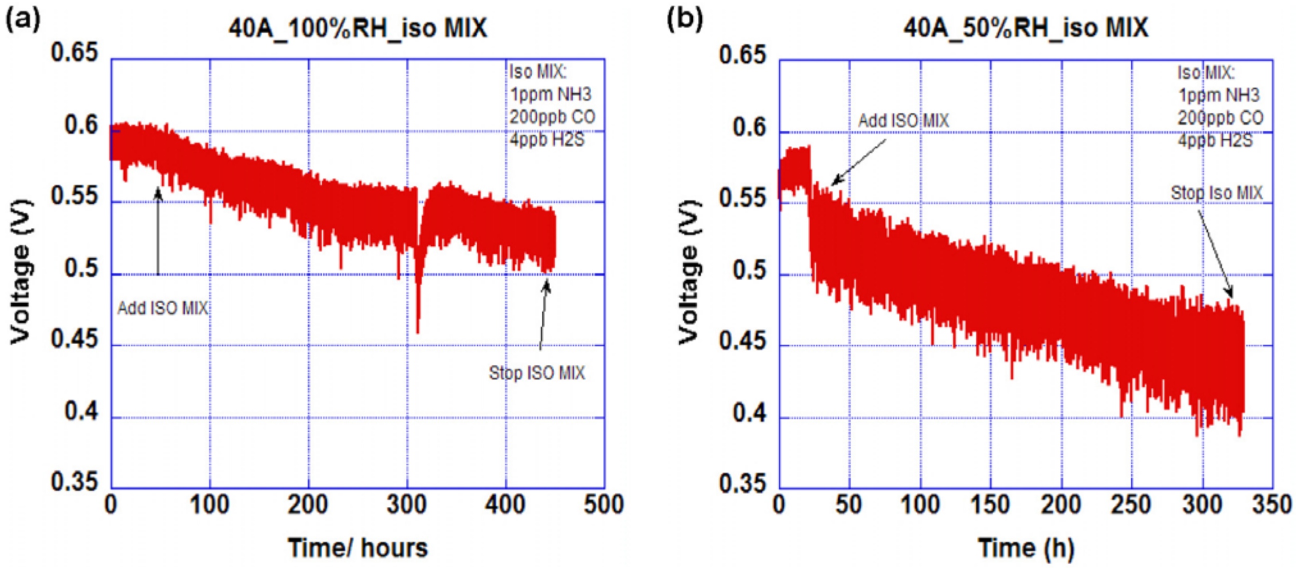

FIGURE 3. LANL MEA tested with 4 ppb H_2S, 100 ppb NH_3, and 200 ppb CO at 80°C in a fuel cell operating at 0.8 A/cm^2 with (a) 50% RH and (b) 100% RH.

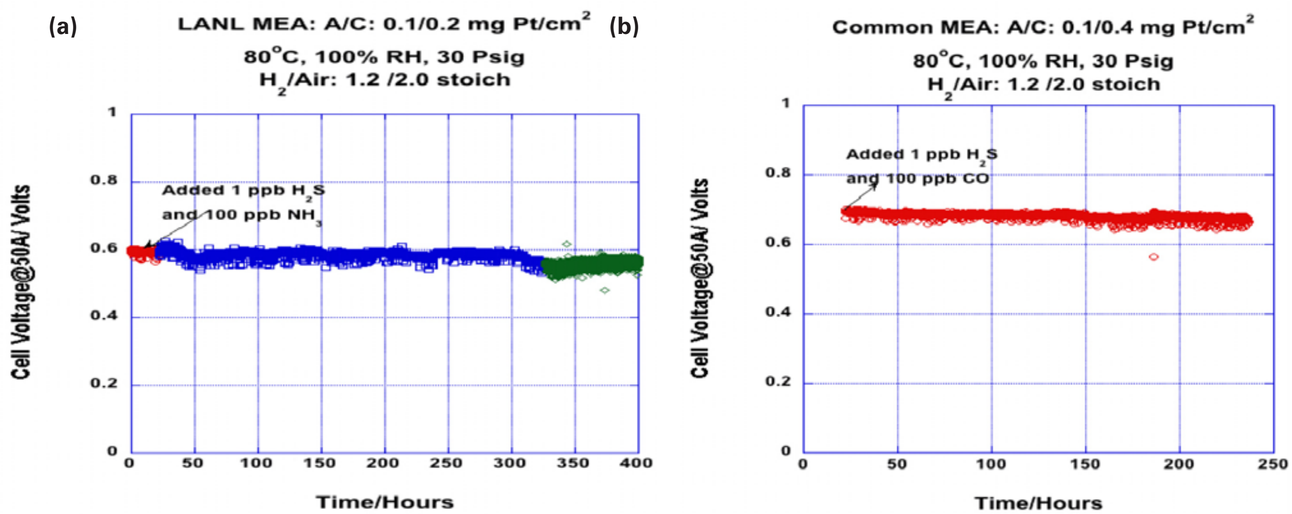

FIGURE 4. Results using the common MEA with 1 ppb H_2S combined with 100 ppb NH_3 (left figure) and 200 ppb CO (right figure) at 80°C for a fuel cell operated at 1 A/cm^2.