Residence Time Distribution of Anode Impurity Pulses in a 47-Cell Commercial PEM Stack

Robert U. Payne, Wenhua H. Zhu, Donald R. Cahela, Amar Tiwari, and Bruce J. Tatarchuk Center for Microfibrous Materiels Manufacturing Department of Chemical Engineering, Auburn University 230 Ross Hall, Auburn, AL 36849, USA brucet@auburn.edu, paynerr@auburn.edu

Concerns over increasing energy consumption coupled with the diminishing supply of fossil-fuels have led to increased research and development in several forms of alternate energy, among which is the polymer electrolyte membrane (PEM) fuel cell. One of the primary obstacles that must be overcome in the development of PEMFC's is the source of hydrogen. PEMFC's are extremely intolerant to impurities, with thresholds of total impurity on the order of .01% [1, 2]. Tolerance level improvement is critical, especially for mobile applications, as it would decrease the mass of the fuel reformer/storage unit and the energy consumed in fuel production. Since PEMFC systems need to be run with higher levels of impurity, the need to understand how these gases behave in the system is apparent. The following work investigates the residence time distribution of inert and reversibly adsorbing impurity pulses introduced into the anode feed of a commercially available PEMFC system.

A 47-cell Ballard[®] Nexa[™] PEM stack was used to investigate the residence time distribution of inert gases (He and Ar) and reversible poisons (CO). Separate voltage taps were attached to each individual MEA of the stack to monitor the time-dependent impurity concentration according to the Nernst equation [2, 3]

$$E = E^{0} + \frac{RT}{2F} \ln \left(\frac{a_{H_{2}} a_{O_{2}}^{\frac{1}{2}}}{a_{H_{2}O}} \right) \text{ which simplifies to } E = E^{0} + \frac{RT}{2F} \ln \left(p_{H_{2}} p_{O_{2}}^{\frac{1}{2}} \right)$$

(where *E* is cell potential; E^0 , standard potential; *a*, activity; and *p*, partial pressure) A septum was installed into the anode feed line just upstream from the stack entrance. The stack was modified in accordance with the work of Zhu et al. to improve the impurity tolerance of the stack by allowing the anode exhaust gas to flow continuously (the system was manufactured to be purged periodically)[1, 3]. A backpressure valve was used in conjunction with a needle valve to regulate the anode outlet (purge) flow rate. The stack was operated using the constant current setting on a TDI programmable load in order to maintain a constant reaction rate. After stack conditions were constant, 5-15 cm³ of impurity gas were injected into the anode feed stream septum using a syringe.

He and Ar were used as inert impurities due to their contrasting diffusivities, while CO was chosen because of its reversibly adsorbing nature and relevance to fossil fuel reformation. Figure 1 shows the combined potential of the two MEA's nearest to the anode outlet, which are fed in parallel. The He (blue) and Ar (red) lines appear similar in shape, but with the more diffusive He exiting the cells sooner than the more sluggish Ar. The CO (green) line is markedly different, because these two cells are fed from the outlets of others; thus, the CO must adsorb and desorb from the catalyst in the previous cells before arriving in the cells under investigation. The collected data were then modeled using residence time distribution theory.

A compartmental model was developed to replicate the RTD data from the impurity pulse experiments, with each cell being modeled using a tanks-in-series model [5]. Figure 2 shows the predicted average concentration in the last cell of the stack, as well as the measured voltage drop of the same cell at approximately 300 W and 500 W stack power. The overall shape of the model approximately agrees with the data, whereas the time-scale does not. While the model necessitates revision, the preliminary results show promise for providing an accurate representation of the time-dependent distribution of impurity gas.

The residence time distribution of various gases injected into the anode feed stream of a PEMFC system were investigated in this work. The significance of the study is in performing stack diagnostics and improving cell layout in order to facilitate impurity tolerance. Manufacturing consistency of large-scale production can also be tested using the described experimental technique. The work also provides a bridge between electrochemical processes and classical chemical reaction engineering.

References

- 1. Ballard Power Systems Inc. *Nexa™ Power Module Installation Manual*. Burnaby, BC: Ballard Power SystemsInc., 2002.
- 2. R. O'Hayre, S. W. Cha, W. Colella, and F. B. Prinz. *Fuel Cell Fundamentals*. New York: John Wiley & Sons Inc., 2006.
- 3. A. J. Bard and L. R. Faulkner. *Electrochemical Methods: Fundamentals and Applications*. 2nd ed. New York: John Wiley & Sons Inc., 1998.
- 4. W. H. Zhu, R. U. Payne, Y. Lu, and B. J. Tatarchuk, "Operation of a PEM Stack with High Impurity Anode Feeds in a Recycle Mode," *2004 AIChE Annual Meeting*. New York: AIChE, 2004.
- 5. O. Levenspiel. *The Chemical Reactor Omnibook*. Corvallis, OR: OSU Bookstores Inc., 2002.



Figure 1: Time-dependent voltage of cells 46 and 47 in a stack at approximately 500 W with 10cm³ pulses of He, Ar, and CO.



Figure 2: Model predictions (red) and experimental data for a 10cm³ He injection at 300 W (green) and 500 W (blue).