Lab Call FY18 (Reversible Fuel Cell): Microstructured Electrodes and Diffusion Layers for Enhanced Transport in Reversible Fuel Cells

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Project Start Date: January 1, 2018 Project End Date: December 31, 2019

Overall Objectives

- Enhance transport performance of unitized reversible fuel cells (URFCs) using arrays of interspersed hydrophobic and hydrophilic channels in electrodes, microporous layers, and gas diffusion layer (GDL) substrates.
- Fabricate, test, and validate URFCs with high performance and durability.

Fiscal Year (FY) 2019 Objectives

- Use characterization tools to determine effect of local microstructure on water management.
- Use diagnostics to determine effect of local microstructures on gas transport.
- Tailor URFC structures based on characterization and diagnostic results to achieve an electrolyzer–fuel cell voltage difference at 0.6 A/cm² of <1.1 V.

Technical Barriers

This project addresses the following technical barriers from the Fuel Cells section of the Fuel

Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan¹:

- (A) Durability
- (B) Cost
- (C) Performance.

Technical Targets

This project focuses on development of materials solutions that can provide enhanced transport and improved performance in URFCs. This approach is expected to meet the following technical targets, which would demonstrate progress toward commercial viability:

- Beginning of life voltage difference between fuel cell mode and electrolyzer mode of less than 1.1 V at 0.6 A/cm²
- Less than 50 mV increase in voltage difference at 0.6 A/cm² after 1,000 cycles between fuel cell and electrolyzer modes.

If PI included a table of technical targets, it should be placed at the beginning of the 1-column section of the report, above the Introduction.

FY 2019 Accomplishments

- Demonstrated preparation of patterned amphiphilic GDL substrates based on localized polytetrafluoroethylene (PTFE) and Nafion deposition.
- Tested performance of URFCs based on patterned GDLs and demonstrated enhanced performance compared to untreated (hydrophilic) and PTFE-treated (hydrophobic) baseline GDLs.

¹ https://www.energy.gov/eere/fuelcells/downloads/fuel-cell-technologies-office-multi-year-research-development-and-22

INTRODUCTION

URFCs, which combine a fuel cell and electrolyzer in a single device [1-4], could address the energy storage challenges associated with the use of intermittent renewable power sources and could be instrumental in realizing the vision of H2@Scale. Several technical barriers must be overcome before URFCs can compete in the marketplace, including the barrier of poor mass transport to and from the oxygen catalyst.

Diffusion media for URFC oxygen electrodes are subject to a wide range of operating voltages, including highly oxidizing conditions approaching 2 V. These high potentials make carbon-based diffusion media unstable during long-term operation, necessitating the use of oxidatively stable materials such as Ti. Diffusion media based on Ti can provide durable operation, but performance lags behind that of carbon-based materials. Furthermore, the wide range of liquid water saturation during URFC operation makes optimization of wettability challenging. New approaches to tailoring materials properties and wettability are needed to enable high-performance operation in both fuel cell and electrolyzer modes

APPROACH

Simultaneously maximizing fuel cell and electrolyzer performance, as well as maintaining performance during repeated mode cycles, are key challenges that can only be addressed by providing fast and effective transport in both fuel cell and electrolyzer modes. Addressing the transport problem is especially difficult because of the conflicting water management requirements: electrolyzers perform best with high liquid water saturation, whereas fuel cells perform best when liquid water saturation is as low as possible while still maintaining effective ionomer hydration. This project addresses transport problems in URFCs through implementation of novel electrode and transport layer microstructures that segregate different transport functionality into different structures. By providing separate hydrophobic and hydrophilic transport pathways, high performance can be achieved in both fuel cell and electrolyzer mode operation.

RESULTS

We prepared patterned hydrophobic/hydrophilic GDL substrates based on PTFE and Nafion segregated in Ti felts (Figure 1) and tested them in operating URFC single cells. Commercial Ti felts were first modified by impregnation with a PTFE dispersion, followed by sintering, yielding a uniformly hydrophobic substrate. Measurement of water contact angles verified the increase in hydrophobicity due to PTFE treatment (Figure 2). Further modification by localized impregnation with Nafion yielded separate hydrophobic and hydrophilic domains in the Ti felt substrate. These separate domains are expected to provide separate transport pathways for gases and for liquid water, respectively.



Hydrophobic Regions (PTFE/Ti)

Figure 1. Scanning electron micrograph cross-section showing localized patterning of a Ti felt substrate to create separate hydrophilic and hydrophobic domains



Figure 2. Water contact angles for Ti felts that are untreated, PTFE-treated, and patterned with PTFE and localized Nafion domains

Diffusion media developed in this project were tested in URFCs to assess the effects of wettability and patterning on performance in both fuel cell and electrolyzer modes. Results shown in Figure 3 provide a direct comparison of performance in fuel cell mode for untreated Ti felts, PTFE-treated Ti felts, and patterned PTFE and Nafion-treated Ti felts. Testing was performed under a variety of hydration conditions ranging from sub-saturated to super-saturated to assess operational robustness under variable conditions. During fuel cell mode testing, we observed membrane electrode assemblies (MEAs) containing untreated Ti felt GDLs to have low performance, with severe flooding occurring at the highest inlet saturation values. Hydrophobic Ti felts prepared using PTFE treatment showed higher performance and greater tolerance to high water saturation, while patterned hydrophilic/hydrophobic Ti felts provided similar or even higher performance.



Figure 3. Fuel cell performance measured using different O₂ electrode diffusion media. Testing was performed at 80 °C, H₂/air, 250 kPa_{abs}. The H₂ electrode used 0.1 mg/cm² Pt/C with a 29BC GDL, while the O₂ electrode used a mixture of 1 mg/cm² Pt and 1 mg/cm² Ir black with Ti felt-based GDLs.

The same URFC MEAs were also tested under electrolyzer mode operation. During electrolysis, we observed MEAs containing untreated Ti felt GDLs and MEAs containing patterned hydrophilic/hydrophobic Ti felts to have similar levels of performance (Figure 4, top). Both these types had higher performance than purely hydrophobic Ti felts, which appeared to cause water starvation. The high hydrophobicity of the PTFE-treated felts reduces liquid water penetration, slowing the rate of water transfer from the flowfield to the O₂ electrode. As a result, the consumption of this limited water supply causes membrane drying, leading to increased high frequency resistance (Figure 4, bottom).



Figure 4. Electrolyzer performance measured using the same MEAs as shown in Figure 3. Testing was performed at 80° C, 75 kPa_{abs}, with liquid water supplied to the O₂ electrode.

The results shown in Figure 3 and Figure 4 demonstrate that our hydrophilic/hydrophobic patterning approach can provide improved performance and improved tolerance to a wide range of hydration conditions in operating URFCs. Using these novel GDL structures, we have demonstrated an electrolyzer–fuel cell voltage difference of 1.1 V at 1.7 A/cm², surpassing the project target of 1.1 V at 0.6 A/cm².

CONCLUSIONS AND UPCOMING ACTIVITIES

Results from FY 2019 have demonstrated the viability of the patterned hydrophilic/hydrophobic diffusion media approach to achieve simultaneous high performance in fuel cell and electrolyzer modes. Further work in FY 2020 will focus on additional tailoring of these structures to increase performance. We will also perform in operando imaging experiments to characterize local water saturation in different types of diffusion media during fuel cell and electrolyzer operation. We will also perform durability testing and work toward achievement of the final project target of 1,000 cycles between fuel cell and electrolyzer mode with less than 50 mV increase in electrolyzer–fuel cell voltage difference at 0.6 A/cm².

FY 2019 PUBLICATIONS/PRESENTATIONS

1. J.S. Spendelow, "New Catalysts and Electrode Structures for Polymer Electrolyte Fuel Cells," Seminar at Case Western Reserve University, February 21, 2019.

REFERENCES

- 1. B. Paul and J. Andrews, "PEM Unitised Reversible/Regenerative Hydrogen Fuel Cell Systems: State of the Art and Technical Challenges," *Renewable and Sustainable Energy Reviews* 79 (2017): 585–599.
- 2. T. Sadhasivam et al., "A Comprehensive Review on Unitized Regenerative Fuel Cells: Crucial Challenges and Developments," *International Journal of Hydrogen Energy* 42 (2017): 4415–4433.
- Y. Wang et al., "A Review on Unitized Regenerative Fuel Cell Technologies, Part-A: Unitized Regenerative Proton Exchange Membrane Fuel Cells," *Renewable and Sustainable Energy Reviews* 65 (2016): 961–977.
- 4. R.J. Remick and D. Wheeler, Reversible Fuel Cells Workshop Summary Report (2011).