Novel Bifunctional Electrocatalysts, Supports, and Membranes for High Performing and Durable Unitized Regenerative Fuel Cells

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- Washington University at St. Louis, St Louis, MO
- Proton OnSite/NEL, Wallingford, CT
- Ballard Power Systems, Burnaby, BC
- Pajarito Powders, Albuquerque, NM

Project Start Date: October 1, 2019
Project End Date: September 31, 2021

Overall Objectives

The goal of this project aims to advance the state of the art of unitized regenerative fuel cells (URFCs) by integrating advanced membrane and ionomer materials with advanced supported bifunctional catalysts into engineered membrane electrode assemblies (MEAs) for URFC operation. The MEAs will be validated for their URFC durability and performance at up to 50 cm^2 active area and validated by Proton and Ballard.

Fiscal Year (FY) 2019 Objectives

- Set and agree upon a common set of baseline materials testing protocols, and execute data management plan.

- Moved out due to subcontracting delays:

  o Synthesize and integrate Pt-Ir catalyst onto Washington University at St. Louis supports, validated with MEA or rotating disk electrode testing, and translate atomic layer deposition synthesized URFC catalysts into an MEA.

  o Demonstrate URFC MEA performance, at the 25 cm^2 level, of 1 A/cm^2 current at >43% round trip efficiency at 80°C and 20 psi balanced pressure in proton exchange membrane fuel cell mode and 20 psi differential pressure in proton exchange membrane water electrolyzer.

Technical Barriers

This project addresses the following technical barriers from the Fuel Cells and Hydrogen Production sections of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration (MYRDD) Plan[1]:

Fuel Cells: Catalyst, Catalyst Support, and MEA:

- A: Durability

- B: Cost

- C: Performance.

Hydrogen Production: Catalyst, Catalyst Support, and MEA:

- F: Capital cost

- G: System efficiency and electricity cost.

Technical Targets

The DOE technical targets and our current project status are listed in Table 1 for comparison.

This project is focused on developing and integrating advanced materials for the membrane and catalyst in order to achieve a highly efficient URFC in constant gas mode:

- Cost: capital cost reduction through reduction in catalyst loading and membrane thickness

- Efficiency improvement: through optimized operating conditions and modes.

FY 2019 Accomplishments

- Set up four subcontracts by June 2020, and kicked off project July 1, 2019, with a 9-month no cost extension.

- Agreed upon a baseline and advanced baseline material set.

- Fabricated and tested baseline electrolysis, fuel cell, and URFC MEAs at Proton OnSite and Lawrence Berkeley National Laboratory (LBNL).

Table 1. Project Technical Targets

Specification	Baseline Fuel Cell	Baseline Electrolyzer	Baseline URFC	Proposed URFC
Membrane thickness (microns)	12	90	125–175	15–30
Total cell Pt loading (mg/cm²)	0.3	1	6	0.55
Ir catalyst loading (mg/cm²)	n/a	2	4	0.6
Fuel cell stack efficiency (%)	>40% (@ 2 A/cm²)	n/a	<40%	>63% (@ 1 A/cm²)
Electrolysis stack efficiency (%)	n/a	>60% (@ 2 A/cm²)	<55%	>80% (@ 1 A/cm²)
Round trip efficiency (%)	n/a	n/a	<25%	>50% (@ 1 A/cm²)
Durability	5,000 auto 28,000 bus	50,000 steady state	n/a	100 h cycling URFC 1000 h discrete <100 μV/h

INTRODUCTION

The Energy Conversion Group at LBNL leads the project effort in developing URFCs (Figure 1), alongside partners Washington University at St. Louis, Proton OnSite, Ballard Power Systems, and Pajarito Powder. URFCs are energy conversion and storage devices that can act in electrolyzer or fuel cell mode in one device, and they represent a promising technology for economic long-term energy storage beyond batteries owing to a combination of high current density and scalable energy storage in the form of hydrogen. The current cost of proton exchange membrane technology makes the business case for discrete systems challenging, except for very long storage times. One approach to reducing the cost is to combine the electrolyzer and fuel cell stacks into a single unitized (URFC) device to reduce material usage. In the past, the much thicker membranes used in electrolyzer stacks made operating them as fuel cells impractical due to the much lower efficiency. Also, platinum is a very poor oxygen evolution catalyst, such that catalyst layers optimized for fuel cells cause significant penalties in electrolyzer mode and vice versa.
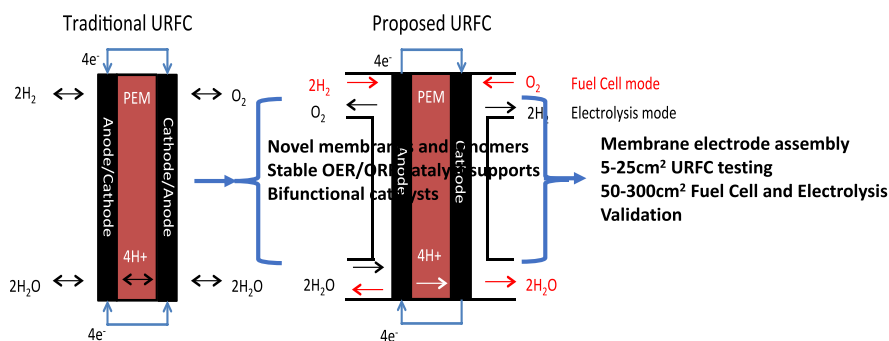

Figure 1. Concept summary with task breakdown

APPROACH

Our approach is to utilize advancements in electrolyzer components to incorporate thin membranes suitable for differential pressure operation (low gas crossover) combined with bifunctional oxygen catalysts on stable engineered supports to demonstrate the feasibility of achieving exceptional efficiencies in both modes. The advanced membrane and supported bifunctional catalysts will be integrated into an MEA that will be

characterized at the subscale in discrete and cycling modes before validation by Proton and Ballard, the industrial leaders in electrolysis and fuel cells.

As URFCs straddle between fuel cell and electrolyzer technology, this project brings industrial experts in both discrete technologies Ballard (Knights, Bashyam) and Proton (Ayers, Capuano) as well as catalyst development (Pajarito Powder, Zulevi). These are well-established leaders in the field of electrolyzers and fuel cells who have the expertise and know-how to integrate and evaluate the URFC MEAs and work alongside fundamental fuel cell and electrolyzer experts at LBNL (Danilovic, Weber) and Washington University at St. Louis (Ramani). Thus, the assembled team is uniquely positioned to advance the state of the art of URFCs as outlined below.

RESULTS

Baseline MEAs were procured from Pajarito Powder as a supplier (not under subcontract yet). They fabricated baseline fuel cell, electrolyzer, and URFC MEAs, which were distributed to Proton and LBNL for testing (using materials in Table 2). The results are shown in Figures 2–4 for 25 cm^2 active area cells. While there were issues with testing the electrolyzer MEA at LBNL, Proton was able to test both the electrolyzer and URFC MEAs and found good agreement in electrolyzer performance even with the reduced Ir loading. Comparing the URFC curves from Proton and LBNL, the data are in reasonably good agreement, within 50 mV of each other. The durability of both the electrolyzer and URFC MEAs at 1 A/cm^2 was very good. The fuel cell performance was not great, and we are still trying to identify the issue that arose with the electrolyzer test as well. Moving forward we will focus on the advanced catalyst configurations.

Table 2. Baseline Configurations

| Material	Fuel Cell MEA	Electrolyzer MEA	URFC MEA
Oxygen reduction reaction catalyst	0.3 mg/cm^2 Pt/C	n/a	1.0–1.5 mg/cm^2 high-surface-area Pt
Hydrogen oxidation reaction catalyst	0.3 mg/cm^2 Pt/C	n/a	0.3 mg/cm^2 Pt/C
Oxygen evolution reaction catalyst	n/a	1.5 mg/cm^2 IrOx	1.5 mg/cm^2 high-surface-area IrOx
Hydrogen evolution reaction catalyst	n/a	0.3 mg/cm^2 Pt/C (TKK)	0.3 mg/cm^2 Pt/C (TKK)
Membrane	Solvay 50 μm-boiled	Solvay 50 μm-boiled	Solvay 50 μm-boiled
Gas diffusion layer	Sigracet 29BC	Sigracet 29BC	Sigracet 29BC
Porous transport layer	n/a	Sintered titanium particles (Mott)	Sintered titanium particles (Mott or Bekeart)
Flowfield cathode	Serpentine graphite	Serpentine graphite	Serpentine graphite
Flowfield anode	Serpentine graphite	Parallel channel titanium	Parallel channel titanium
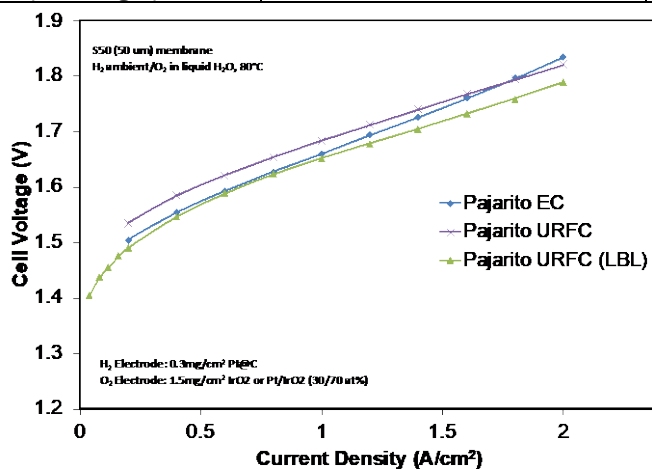
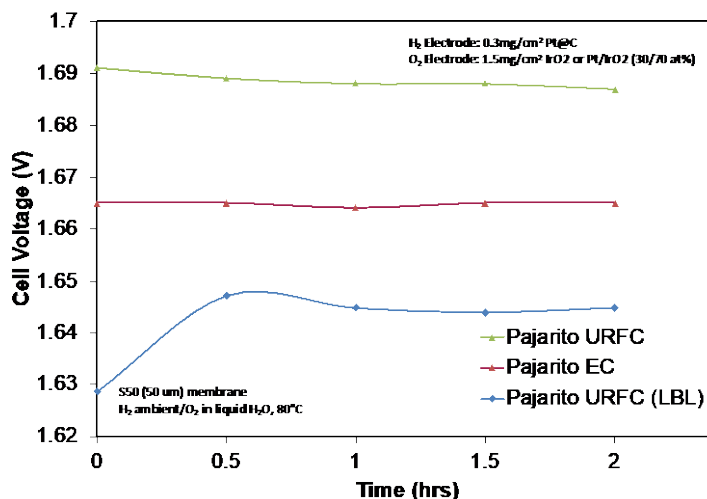Figure 2. Electrolyzer and URFC MEA baseline measurements

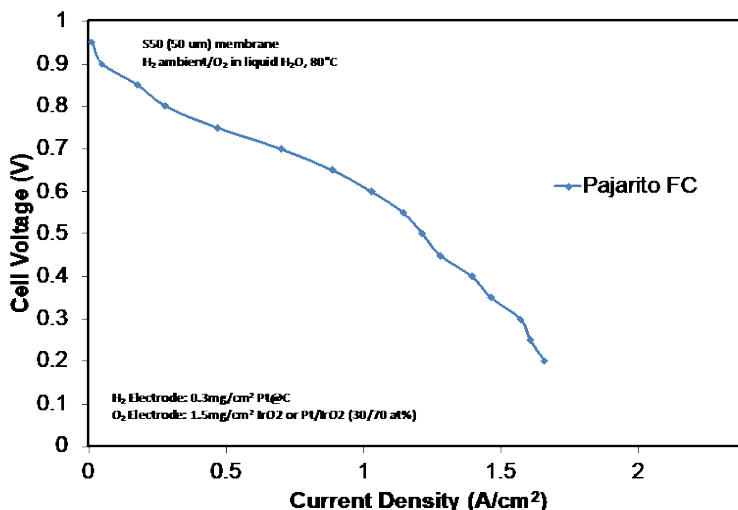Figure 3. Electrolyzer and URFC MEA durability

Figure 4. Fuel cell baseline polarization curve

Proposed Configuration for Integration and Go/No-Go Milestone

Due to subcontracting delays and in order to attempt to maintain the project schedule, we will shift out milestones by 9 months. We will continue using the baseline Solvay 50 mm membrane for development of the MEA, which will incorporate advanced baseline catalysts. These will be materials that Pajarito has that are either high-surface-area unsupported Pt and Ir, or Pt and Ir supported on TiO_2, both of which will better approximate the behavior of the Washington University at St. Louis supported catalysts.

Testing Protocols

The accelerated stress tests (ASTs) will consist of our proposed URFC AST, membrane durability, and a stretch goal of performing the catalyst, support, and membrane durability tests if time and budget allow:

- URFC AST: a fuel cell break in followed by fuel cell performance and electrolyzer testing. The anode side is then cycled potentiostatically between 0.55 V and 1.65 (1 A/cm^2 in electrolysis mode) for n number of cycles up to 10,000 with liquid water flowing on the anode during the AST. Periodically the test is stopped in order to test the electrolyzer and fuel cell performance. Ballard will also evaluate

the catalyst and support ASTs and determine how they compare and what the degradation mode is between the three ASTs.

- Fuel cell catalyst durability: as in MYRDD Plan.

- Support durability: as in MYRDD Plan.

- Membrane durability: as in MYRDD Plan.

CONCLUSIONS AND UPCOMING ACTIVITIES

In conclusion, due to subcontracting delays this project is on a 9-month shift in effort. Nevertheless, much was achieved in FY 2019 and there is much to do in the coming years.

Remaining work prior to end of project:

1. Integrate Washington University at St. Louis supports with bifunctional catalysts and demonstrate URFC operation.

2. Down-select commercial or Washington University at St. Louis membranes that can achieve high efficiency and durability under fuel cell and electrolyzer conditions.

3. Determine optimal fuel cell operating conditions to optimize performance.