



Regenerative Fuel Cell System

Cooperative Research and Development Final Report

CRADA Number: CRD-16-635

NREL Technical Contact: Shaun Alia

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Contract No. DE-AC36-08GO28308

**Technical Report
NREL/TP-5900-76132
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Cooperative Research and Development Final Report

Report Date: 1/30/20

In accordance with requirements set forth in the terms of the CRADA agreement, this document is the final CRADA report, including a list of subject inventions, to be forwarded to the DOE Office of Science and Technical Information as part of the commitment to the public to demonstrate results of federally funded research.

Parties to the Agreement: pH Matter, LLC

CRADA number: CRD-16-635

CRADA Title: Regenerative Fuel Cell System

Joint Work Statement Funding Table showing DOE commitment: No NREL shared resources

Estimated Costs	NREL Shared Resources a/k/a Government In-Kind
Year 1	\$0.00
TOTAL	\$0.00

Abstract of CRADA Work:

Grid energy storage is required to maintain reliable electricity delivery from energy producers to their customers. As electrical loads on the grid change throughout the day, stored energy supplies electricity during increased power demand periods. Further, as more renewable and alternative energy sources are added, energy storage will maximize the usefulness of these technologies. Currently, the United States has over 24.6 GW of grid storage, about 2% of the total electrical production capacity. About 95% of energy storage comes from pumped hydro. However, pumped hydro energy storage is dependent on existing geographic features, making it difficult to integrate with widespread renewable energy sources. As energy demands continue to expand, and more renewable energy (i.e. wind and solar) is added to the grid, new energy storage technologies will be needed that are not dependent on geographic features. Battery technologies can provide energy storage for some applications, but are not economically well-suited for long duration charge/discharges, such as load-leveling of renewable energy. Consequently, development of new energy storage devices will augment the existing grid and reduce the capital investment in construction upgrades. As ever-increasing mandates for renewable energy are implemented, lower-cost energy storage solutions for renewable energy will be necessary to keep electricity costs low for consumers.

Regenerative fuel cells offer a unique solution for grid energy storage. Unlike batteries, regenerative fuel cells can cost-effectively store a large amount of energy in the form of hydrogen. Energy in the form of hydrogen can be stored at a cost of about \$35/kW-hr, versus \$196/kW-hr for batteries. The regenerative fuel cell systems could also provide an added benefit of hydrogen generation for fuel cell vehicles. Regenerative fuel cells typically operate in either acidic (i.e. PEM fuel cells) or alkaline environments. The relevant reactions for regenerative fuel cells are shown below:

(1) Oxygen Reduction Reaction (ORR) in acid: $O_2 + 4 H^+ + 4 e^- \rightarrow 2 H_2O$

- (2) Oxygen Reduction Reaction in alkaline: $O_2 + 2 H_2O + 4 e^- \rightarrow 4 OH^-$
- (3) Hydrogen Oxidation Reaction (HOR) in acid: $2 H_2 + 4 e^- \rightarrow 4 H^+$
- (4) Hydrogen Oxidation Reaction in alkaline: $2 H_2 + 4 OH^- \rightarrow 4 H_2O + 4 e^-$
- (5) Overall Energy Generation Reaction: $2 H_2 + O_2 \rightarrow 2 H_2O$
- (6) Overall Energy Storage Reaction: $2 H_2O \rightarrow 2 H_2 + O_2$

The overall reactions are the same in acid or alkaline, the only difference is the ions being exchanged. The Oxygen Evolution Reaction (OER) and Hydrogen Evolution Reaction (HER), which occur during energy storage modes, are the reverse of the ORR and HOR, respectively.

Summary of Research Results:

Task 1.1. Low-PGM Anodes (NREL Lead). NREL developed low-platinum group metal (PGM) catalysts as part of this project, activity in the hydrogen oxidation (HOR) and evolution reactions (HER). A focus of this task was improving activity beyond non-PGM materials while keeping the PGM content low to minimize catalyst layer cost. Spontaneous galvanic displacement was used to coat nickel (Ni) nanowires with platinum (Pt) layers. Initial performances in half-cell tests were found to produce consistent site-specific activities across a range of compositions. Mass activity improved at lower displacement levels likely due to increased Pt utilization with thinner layers (Figure 1).

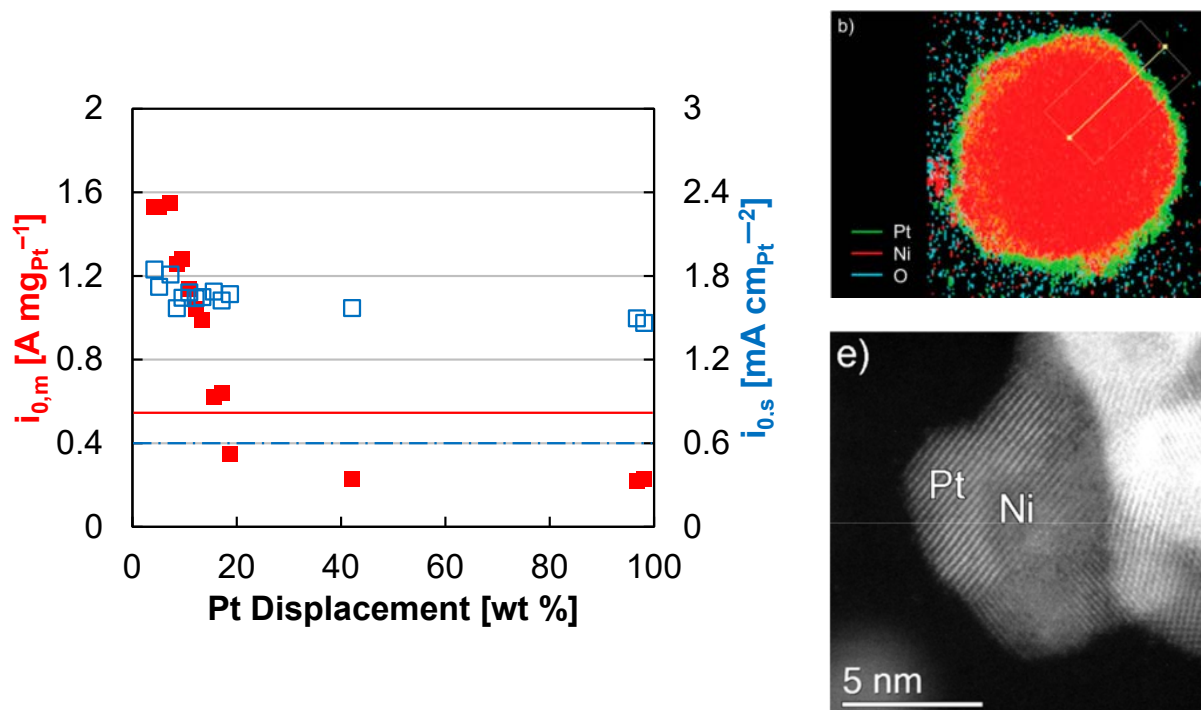


Figure 1. (a) HOR/HER activity of Pt-Ni nanowires as a function of composition. Microscopy of Pt-Ni nanowires (b) cross-sectioned and (c) focused on Pt and Ni lattices and zone integration.

Subsequent optimization of Pt-Ni nanowires focused on hydrogen annealing to improve Pt/Ni zone integration. Pt lattice compression was found to improve activity, attributed to lattice compression

weakening Pt-H binding. Optimized Pt-Ni nanowires demonstrated significant benefits to as-synthesized nanowires and baseline commercial catalysts (Figure 2).

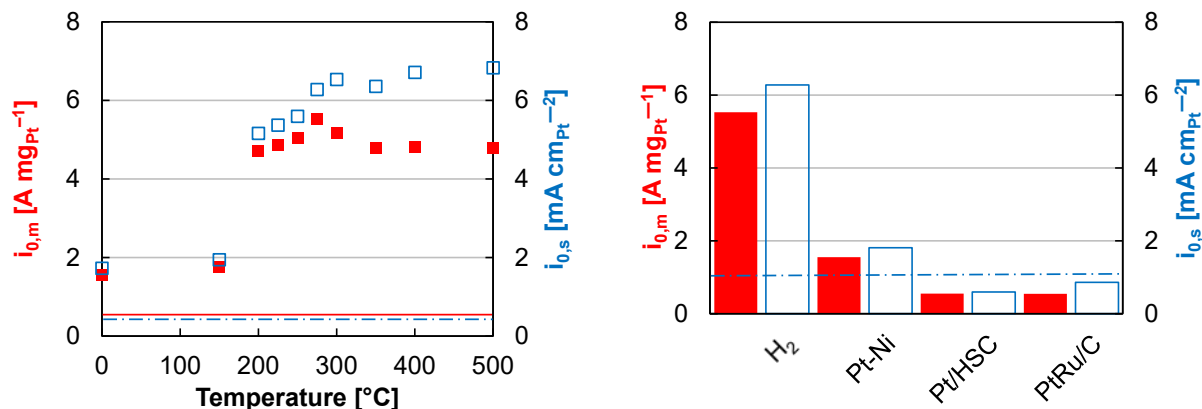


Figure 2. (a) HOR/HER activity of Pt-Ni nanowires as a function of hydrogen annealing temperature. (b) HOR/HER activity of hydrogen annealed (H₂) and as-synthesized (Pt-Ni) nanowires compared to commercial baselines (Pt/HSC, PtRu/C).

Task 1.2. Non-PGM Anodes (pHM Lead). This task was completed by pHM.

pHM developed a CN_xP_y catalyst, COR-2, as a PGM-free option in fuel cell anodes. Testing in fuel cell and electrolyzer modes showed promising performance, with small performance losses compared to PtRu baselines (Figure 3).

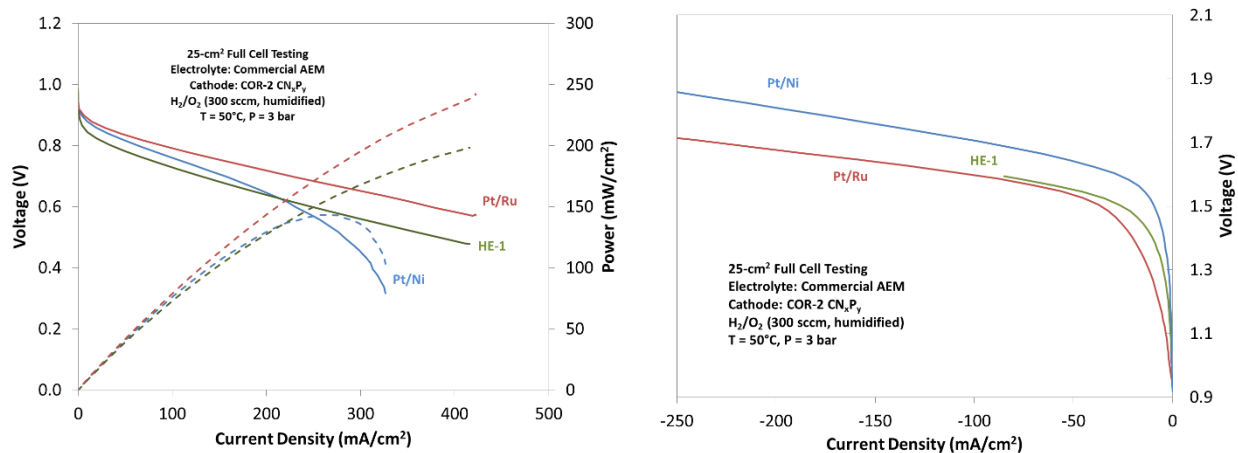


Figure 3. MEA performance of HE-1, Pt/Ru, and Pt/Ni in (a) fuel cell and (b) electrolyzer modes.

Task 1.3. Anode Scale-up (pHM and NREL). Pt-Ni nanowires were down-selected to the high-performer following the hydrogen annealing step. Catalyst synthesis was scaled to approximately 400 mg using galvanic displacement. Atomic layer deposition was also used to increase yield to a 10 g batch size. These materials were delivered to pH Matter for cell level implementation and testing. Materials scale up of down-selected low-PGM hydrogen anode catalysts supported larger cell and stack tests (**Objective 2, Task 4.1**).

Task 2.1 MEA Synthesis (All Members). Catalysts from Task 1 were used for MEA fabrication. The following parameters were investigated:

- 1) Anion-exchange ionomer and membranes categories: Nafion, Tokuyama, and NREL PFAEM (Gen 2) ionomers were used this project.
- 2) Solvents: Isopropanol and water were used as solvents.
- 3) Anion-exchange ionomer loading was optimized to improve cell performance.
- 4) NREL used spraying onto GDEs and PTLs to evaluate performance and durability.

Following examination of the above parameters, NREL PFAEM at an ionomer to catalyst ratio of 0.2 was used for cell testing (Figure 4). These specifications resulted in optimal performance in electrolyzer testing. Spraying onto GDEs was necessary to ensure stable performance.

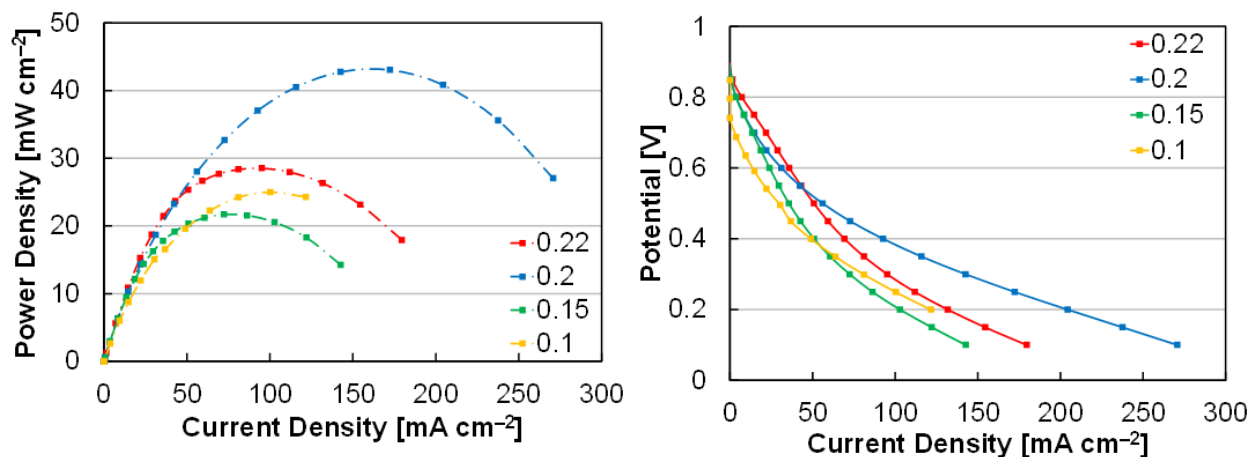


Figure 4. MEA performance of NREL Gen 2 membranes in (a) fuel cell and (b) electrolyzer modes, varying ionomer content.

Task 2.2. Cathode Optimization for Higher Temperature and Pressure (pHM Lead). This task was completed by pHM.

Task 2.3. GDE Scale-up (pHM Lead). This task was completed by pHM.

pHM scaled the synthesis of HE-1 and COR-2 and sprayed GDEs to provide other project partners (NREL, Giner) for cell testing in Task 3.

Task 3.1. Performance Testing (All). As part of the performance testing task, NREL provided membrane and ionomer samples (PFAEM Gen 2) to pH Matter and Giner.

NREL cell testing used a 25-cm² cell test stand to obtain I-V curves, demonstrate steady state performance for ORR and OER, and initial cycling stability under baseline conditions. Cell testing of materials included oxygen (COR) and hydrogen (HyROC) electrode catalysts provided by pH Matter and baseline catalysts (Pt in hydrogen, Ir in oxygen) in proton exchange membrane-based systems. Initial testing of HyROC/COR showed promising beginning of life performance, roughly one third the performance of Pt/Ir (Figure 5). Initial stability testing showed losses in both systems, found in dropping activity and thinning cyclic voltammograms. To evaluate catalyst loss relative to other system components, performance/durability was repeated for HyROC/COR with Pt/Ti PTLs (Figure 6). Losses in stability testing were significantly less than with carbon PTLs. Losses were also very small following changing the testing electrolyte, indicating that contaminants/carbonation contributed to loss in extended operation. Comparisons of beginning of life and post-durability performance were favorable and little loss was observed for HyROC/COR catalysts.

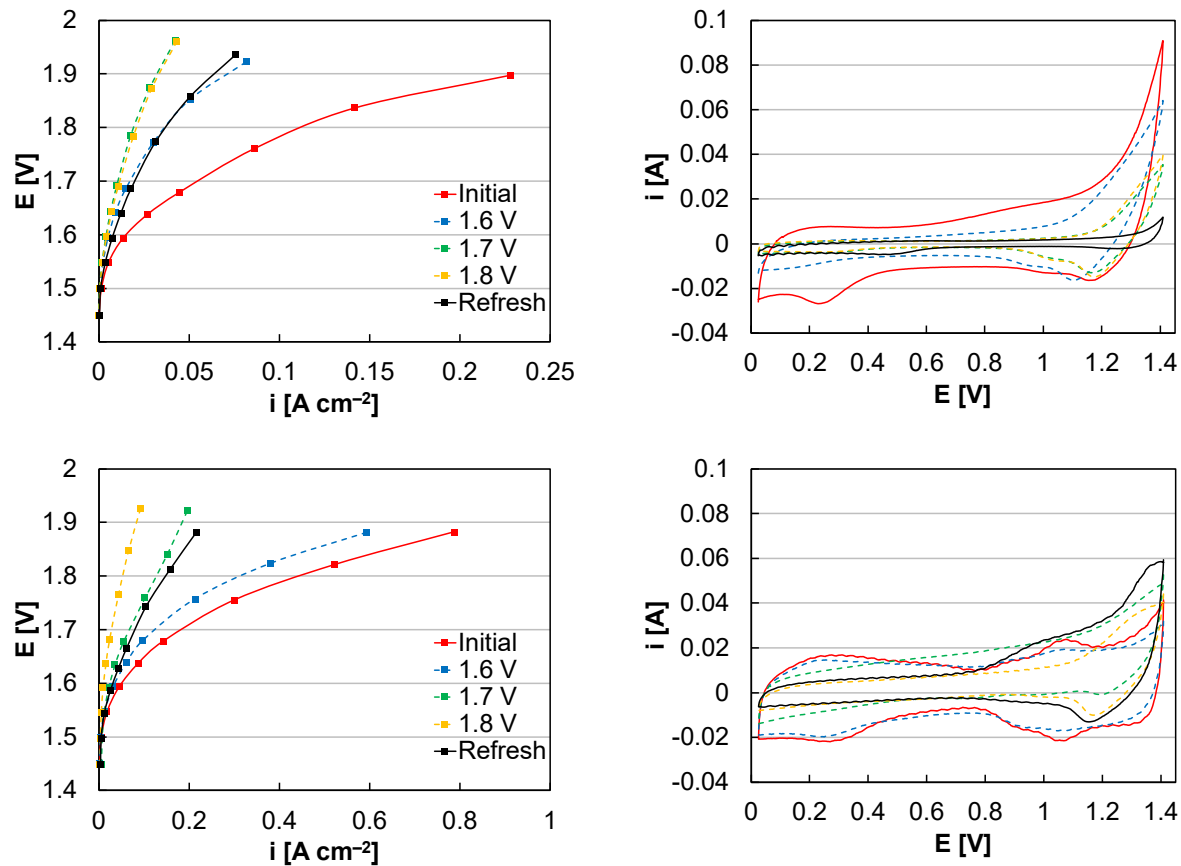


Figure 5. Polarization curves and cyclic voltammograms of MEAs containing (a-b) HyROC/COR and (c-d) Pt/Ir on carbon (Toray) PTLs as a function of stability testing, initially (red) and following a two-day 1.6 V hold (blue), a two-day 1.7 V hold (green), a two-day 1.8 V hold (yellow), and following an electrolyte change (black).

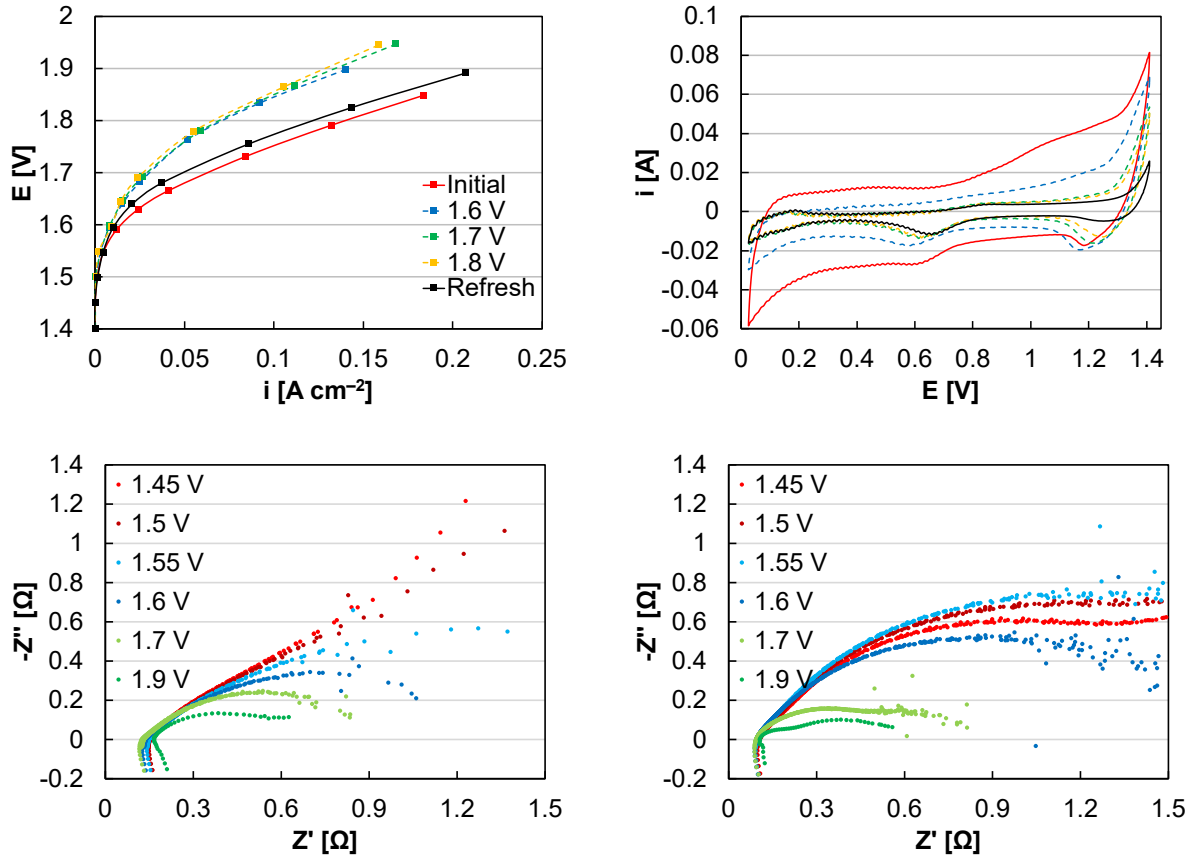


Figure 6. (a) Polarization curves and (b) cyclic voltammograms of MEAs containing HyROC/COR on Pt/Ti PTLs as a function of stability testing, initially (red) and following a two-day 1.6 V hold (blue), a 2 day 1.7 V hold (green), a two-day 1.8 V hold (yellow), and following an electrolyte change (black). Impedance data at (c) beginning of life and (d) the electrolyte refresh after the six-day stability test.

Task 3.2. Load-Cycle Testing (Giner Lead). This task was completed by Giner and pHM.

A MEA single-cell was tested with a simulated operation profile for over 250 h. Under these load-cycle conditions, no losses were found in electrolyzer or fuel cell modes (Figure 7).

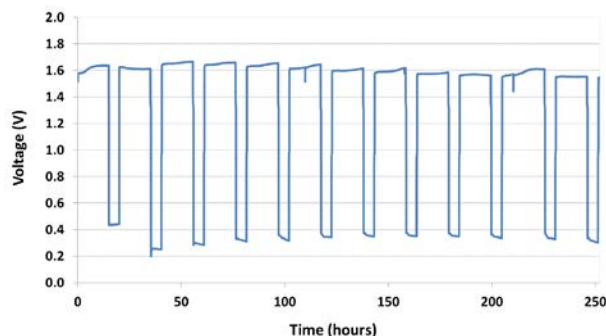


Figure 7. Cell voltage as a function of operation time. The MEA was tested for over 250 h using HE-1 as the hydrogen electrode catalyst and COR-2 as the oxygen electrode catalyst.

Task 4.1. Stack Fabrication (Giner Lead, All support).

Task 4.2. Stack Testing (Giner).

The following text described results from Tasks 4.1 and 4.2. Stack fabrication was completed by Giner and stack testing by pHM.

pHM evaluated a short stack (3 cell) with 3 bar of back pressure at 50°C. The 3-cell (25 cm² each) stack was demonstrated for 200 cycles between 50 mA/cm² (electrolysis mode) and 150 mA/cm² (fuel cell mode) (Figure 8). The cell utilized HE-1 at the hydrogen electrode and COR-2 at the oxygen electrode.

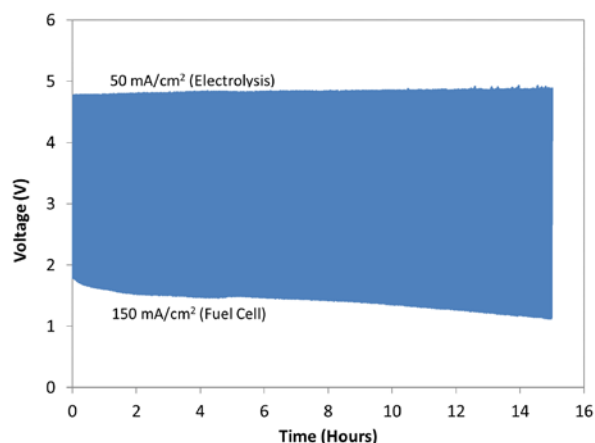


Figure 8. Cell potential as a function of operation time during cycling (200 cycles) between 50 mA/cm² (electrolysis mode) and 150 mA/cm² (fuel cell mode).

Task 5. Economic Modeling (pH Matter Lead, All support).

Task 5.1. Material Scale-up Projections.

Task 5.2. Delivered Electricity Projections.

The following text described results from Tasks 5.1 and 5.2. This task was led by pHM. NREL contributed electrolyzer operation expectations to add to modeling projections.

Deliverable energy costs were determined for fuel cells operating as discrete and unitized systems, where a unitized reversible fuel cell offers a cost benefit in delivered energy cost.

Table 1. Cost analysis of fuel cells/electrolyzers operating as discrete and unitized systems.

	Discrete	Unitized
Upfront Power Cost (\$/kW)	Fuel Cell* + Electrolyzer**	Reversible Cell Stack
Capacity Cost (\$/kW)	\$23***	\$35
RT Efficiency	43%	43%
Delivered Energy (\$/kW-hr) Steward et al. case	0.223	0.175

Task 6. Management and Reporting. NREL contributed to project meetings and reports through bi-weekly teleconferences and quarterly reports.

Subject Inventions Listing:

N/A

ROI #:

N/A

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