

## V.E.1 Regenerative Fuel Cell System

Paul Matter (Primary Contact), Minette Ocampo,  
Michael Beachy, and Chris Holt

pH Matter, LLC  
1275 Kinnear Rd.  
Columbus, OH 43212  
Phone: (614) 484-5038  
Email: info@phmatter.com

DOE Manager: Donna Ho

Phone: (202) 586-8000  
Email: Donna.Ho@ee.doe.gov

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Subcontractors:

- Giner, Inc., Newton, MA
- National Renewable Energy Laboratory (NREL), Golden, CO

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### Technical Barriers

This project addresses the following technical barriers from the Fuel Cell section of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan, with respect to AEMFCs for energy storage:

- (A) Durability: increase the durability/stability of catalysts
- (B) Cost: development of low-cost PGM-free catalysts for reversible anion-exchange membrane fuel cells
- (C) Performance: integrate catalysts with membranes and gas diffusion layers into MEAs that operate at high power and efficiency

### Technical Targets

This Phase II Small Business Innovation Research project is developing new catalyst materials and MEAs for a regenerative AEMFC stack. The materials being developed address the following technical targets for energy storage applications:

- 1,000 cycles above target operating efficiency and current density
- 42% efficiency; >250 mA/cm<sup>2</sup> power generation; >50 mA/cm<sup>2</sup> energy storage

### FY 2016 Accomplishments

Since the previous reporting period, the following work related to the technical objectives has been accomplished on this Small Business Innovation Research Phase I and Phase II project:

- In half-cell testing, demonstrated GDE for 200 cycles between projected oxygen reduction reaction (ORR) and oxygen evolution reaction (OER) voltages at 50°C, and ORR current density of 200 mA/cm<sup>2</sup>.
- Built an economic model as a basis for the technical targets in the Phase II project. The model indicated that if the performance targets are achieved at the system level, then the reversible AEMFC could deliver electricity at <\$0.18/kWh using the assumptions developed by Steward et al. [1].
- Established baseline MEA performance in AEMFC single cell testing.
- Began evaluation of PGM-free hydrogen electrode materials.

### Overall Objectives

- Demonstrate a reversible 25 cm<sup>2</sup> anion exchange membrane fuel cell (AEMFC) for 1,000 cycles (42% round-trip efficiency; >250 mA/cm<sup>2</sup> power generation; >50 mA/cm<sup>2</sup> energy storage).
- Incorporate membrane electrode assemblies (MEAs) into a regenerative stack.
- Perform economic analysis on reversible AEMFC system following established DOE guidelines for candidate grid load leveling technologies.

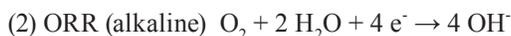
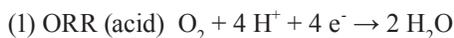
### Fiscal Year (FY) 2016 Objectives

- Prepare and characterize a matrix of precious group metal (PGM) free catalysts and incorporate them into MEAs for use in reversible AEMFC systems.
- Demonstrate a bi-functional gas diffusion electrode (GDE) that is consistent with DOE AEMFC performance targets with <10% degradation over hundreds of cycles.
- Perform economic analysis on a reversible AEMFC system following established DOE guidelines for candidate grid load leveling technologies.



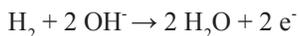
## INTRODUCTION

Low temperature fuel cells, such as proton exchange membrane (PEM) and AEMFCs, offer an efficient and clean means of energy conversion of hydrogen to electricity. However, PEM fuel cells typically require platinum in the cathode to operate at high power density and high efficiency, which hurts the economics for this technology. Platinum is used as an electro-catalyst for the ORR, the cathode side half reaction is shown below for acidic and alkaline electrolytes, respectively:



The slow kinetics in the cathode is one of the largest sources of inefficiency in fuel cells, thus high platinum catalyst loadings are needed to prevent even more voltage losses (or overpotential). At commercial scale, precious metals in the cathodes of PEM fuel cells would comprise a significant portion of the entire stack cost [1,2]. Additionally, Pt-based ORR catalysts can degrade quickly under fuel cell operating conditions, such as frequent load cycling.

More recently, there has been renewed interest in AEMFCs for stationary applications. Development of commercial anion exchange membranes is helping to alleviate system-level problems with alkaline fuel cells, such as pressure balance. Further, recent published results at Los Alamos National Laboratory have shown that alkaline fuel cells could potentially operate at high efficiency with non-platinum ORR catalysts [3]. AEMFCs are of particular interest for energy storage applications that do not have size or volume limitations, such as grid load leveling. In an alkaline fuel cell oxygen is reduced by reaction (2) above, and hydrogen is oxidized by reaction (3) below.



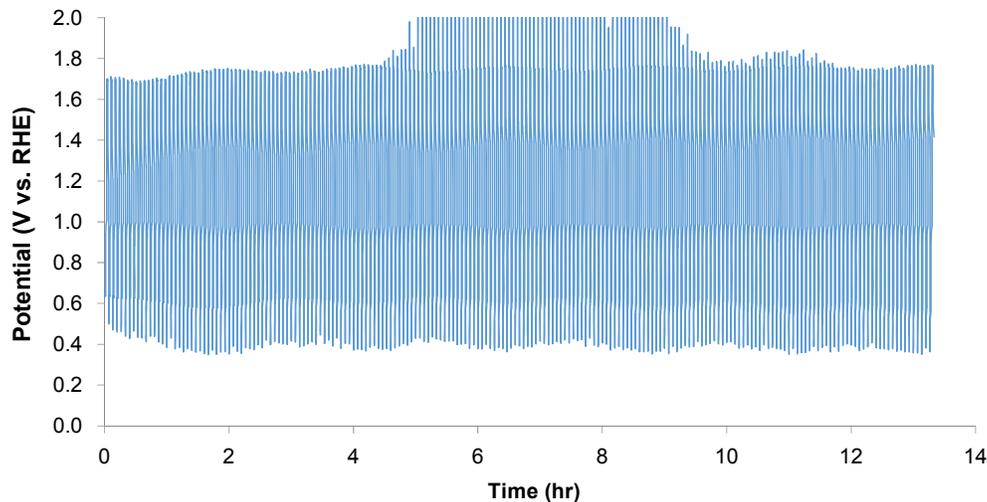
AEMFCs could potentially be operated in a reversible manner, allowing renewable energy to be stored in the form of hydrogen. This would be particularly valuable when coupled with renewable energy generation (wind or solar) to provide energy storage and load leveling. However, when operating in regeneration mode, cathode degradation is even more pronounced for conventional ORR catalysts because of the high voltages required for the OER, the reverse of Reaction 2 above. Consequently, in existing reversible systems, separate cell stacks for fuel cell and electrolysis operation are used, adding to the already high system cost. If a low-cost regenerative stack could be developed, it would be a key breakthrough in the commercial viability of energy storage systems [4]. In this project, pH Matter, LLC is partnering with Giner, Inc., and NREL to develop and demonstrate a low-cost regenerative AEMFC system.

## APPROACH

The overall objective of the proposed project is to develop and demonstrate a regenerative fuel cell stack technology that is economically viable in stationary energy storage. In the project, researchers at pH Matter will synthesize a matrix of PGM-free HOR/hydrogen evolution reaction (HER) catalysts, and GDEs based on these materials. Researchers at NREL will synthesize a matrix of low-PGM hydrogen electrode materials. The HOR/HER materials and GDEs will be fully characterized and tested under cycling conditions to determine performance and stability. Additionally, pH Matter will further optimize nitrogen- and phosphorus-doped carbon electrodes ORR/OER electrodes developed in Phase I for improved performance and durability at higher temperatures and pressures. The hydrogen and oxygen electrodes will then be demonstrated in 25 cm<sup>2</sup> single cells for over 1,000 cycles. Cells that degrade during cycling will be characterized by pH Matter and NREL to determine degradation mechanisms. This information will be used to iteratively prepare more optimized cells. Engineers at Giner will test cells in conjunction with Giner's water-management membrane technology. Down-selected cells will then be incorporated into a regenerative fuel cell stack and demonstrated in simulated application testing at Giner. The project will establish a foundation for future work, where the technology will be incorporated into a prototype regenerative fuel cell system. Additionally, a design and economic model of the regenerative fuel cell system will be built to verify advantages of the approach compared to available energy storage technologies. The successful result of the proposed Phase II work will demonstrate the feasibility of a regenerative fuel cell system with economic advantages compared to existing technologies.

## RESULTS

In previous Phase I work on this project a matrix of novel PGM-free catalysts for ORR and OER were synthesized and tested in a rotating disk electrode set-up. GDEs were made using a screen-printing method using down-selected catalysts. Various catalyst formulations, catalyst loadings, ionomers and/or binders, ink compositions, and electrode substrates were examined. Testing was conducted with commercial AEMs in an in-house constructed stainless steel half-cell set-up. For the grid load-leveling application, it is expected that current density will be highest (by a factor of 5–6) during periodic cell discharges (ORR operation) compared to OER operation. Half-cell GDE testing examined cycling between ORR and OER conditions. For these tests at 45°C, cycles were conducted at 40 mA/cm<sup>2</sup> for OER, and 200 mA/cm<sup>2</sup> for ORR with the direction of the current being reversed every 2 min (1 min of current, 1 min of rest). Some GDE configurations showed excellent stability for ORR and OER cycling during these tests in up to 200 cycles. Figure 1

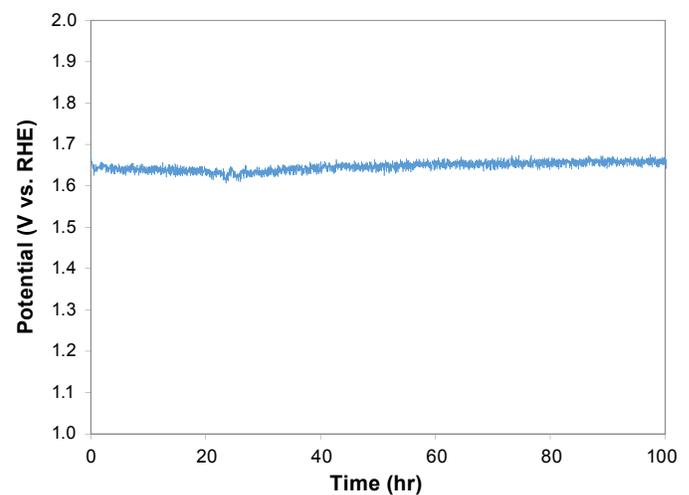


**FIGURE 1.** ORR-OER cycle testing for 200 cycles obtained in half-cell GDE at 50°C for non-precious metal catalyst in humidified oxygen; aqueous 5 M KOH fed to the counter electrode chamber; 200 mA/cm<sup>2</sup> ORR, 50 mA/cm<sup>2</sup> OER

shows the half-cell cycle test for pH Matter’s non-PGM GDE compressed to a commercial AEM, and operating in pure oxygen. The results demonstrate the ability of this class of catalysts to undergo a number of cycles without rapid degradation.

Testing also examined long-term durability of electrodes at steady-state for OER or ORR. In testing on parallel projects, the stability of the non-PGM catalysts were found to be excellent over 100 h at 70°C during steady-state ORR operation at 350 mA/cm<sup>2</sup> in pure oxygen at 1 atm (data not shown). No degradation could be measured, as performance improved slightly over a 100-hour test. Stability was also excellent for steady-state OER testing over 100 h, as shown in Figure 2. For operation of the non-PGM GDE, the electrode was pressed to a commercial AEM and flooded with 5 M KOH at 45°C and run at 40 mA/cm<sup>2</sup>. The OER stability was excellent over the 100-hour test.

At the end of the Phase I Small Business Innovation Research, an economic model was updated to project electricity costs for energy stored with a reversible AEMFC system based on test results. The guidelines for the model and assumptions generally followed those used by Steward et al. [1], but assumed a reversible AEMFC stack that could operate at Phase II targets. The model demonstrated that if technical targets can be achieved at the stack scale, then a reversible AEMFC would be cost-competitive with compressed air energy storage and pumped hydro energy storage approaches. However, unlike these approaches, a fuel cell system is not subject to geologic restrictions. The projected delivered electricity would cost less than \$0.18/kWh. The model was also used to determine sensitivity of the electricity cost to a number of factors, particularly those that have yet to be demonstrated. The sensitivity analysis found



**FIGURE 2.** Steady-state OER testing over 100 h at 45°C for non-precious metal catalyst in 5 M KOH operating at 40 mA/cm<sup>2</sup> OER

that competitive economic performance will be dependent on achieving a stack life-time of greater than four years.

In the first quarter of the Phase II project, work has begun on development of non-PGM hydrogen electrode catalysts for the HOR and HER. Testing has focused on obtaining baseline performance and stability for platinum–ruthenium catalysts, and comparison to non-PGM catalysts. The durability during cycling for the hydrogen electrode appears to be less challenging than the oxygen electrode, as all materials tested have shown excellent durability. However, further performance improvements are required to match the PGM standard. Further optimization has also begun on the non-PGM ORR/OER electrodes. Future work will target

cycling and performance demonstration in single cells with the non-PGM electrodes.

## CONCLUSIONS AND FUTURE DIRECTIONS

The following conclusions can be drawn from work completed to this point:

- The novel ORR/OER PGM-free catalysts being developed in this project show performance in alkaline conditions comparable to that of precious metal ORR/OER catalysts, and good stability during cycling from ORR to OER voltages.
- ORR/OER cycling stability for 200 cycles has been demonstrated with GDEs, and degradation during steady-state operation is less of a concern.
- Economic modeling suggests that the reversible AEM fuel cell concept would be an excellent energy storage option for grid load leveling if performance targets can be achieved at the system level.

Future work in the remainder of the Phase II project will include:

- Further hydrogen electrode materials development and incorporation of the materials into cells and stacks.
- Optimization of the electrode-membrane interface.
- Demonstration of low-cost 25 cm<sup>2</sup> reversible cells.
- Characterization of the electrodes before and after cycling to better understand any degradation mechanisms.
- Demonstration of single cell durability over 1,000 cycles.
- Demonstration of a regenerative stack.
- Design of a prototype energy storage system that incorporates the stack.
- Economic analysis of a reversible AEMFC system for a specific energy storage application.

## FY 2016 PUBLICATIONS/PRESENTATIONS

1. Matter, Paul H., Minette Ocampo, Michael Beachy, and Chris Holt, "Regenerative Fuel Cell System," *DOE Hydrogen and Fuel Cells Program Annual Merit Review and Peer Evaluation* (2016), June 8, 2016, Washington, D.C.

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2. James, Brian D., Jennie M. Moton, and Whitney G. Colella, "Mass Production Cost Estimation of Direct H<sub>2</sub> PEM Fuel Cell Systems for Transportation Applications: 2013 Update." Accessed July 23, 2015. [http://energy.gov/sites/prod/files/2014/11/f19/fcto\\_sa\\_2013\\_pemfc\\_transportation\\_cost\\_analysis.pdf](http://energy.gov/sites/prod/files/2014/11/f19/fcto_sa_2013_pemfc_transportation_cost_analysis.pdf)
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