# V.A.7 Non-Precious Metal Fuel Cell Cathodes: Catalyst Development and Electrode Structure Design

Piotr Zelenay (Primary Contact), H. Chung, U. Martinez, E. Holby, U. Tylus, G. Purdy Materials Physics and Applications Division Los Alamos National Laboratory (LANL) Los Alamos, NM 87545 Phone: (505) 667-0197, Fax: (505) 665-4292 Email: zelenay@lanl.gov

#### DOE Manager

Nancy Garland Phone: (202) 586-5673 Email: Nancy.Garland@ee.doe.gov

#### Subcontractors

- J. Ziegelbauer (PI), General Motors, Warren, MI
- M. Odgaard (PI), D. Schlueter, IRD Fuel Cells, Albuquerque, NM
- S. Litster (PI), S.K. Babu, Carnegie Mellon University (CMU), Pittsburgh, PA
- M. Neidig (PI), J. Kehl, J. Kneebone, University of Rochester, Rochester, NY
- Z. Chen (PI), D. Higgins, G. Jiang, M.-H. Seo, University of Waterloo, Waterloo, Canada
- K.L More (PI), D. Cullen, Oak Ridge National Laboratory (ORNL), Oak Ridge, TN

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# **Overall Objectives**

- Advance non-platinum group metal (PGM) cathode technology through the development of new materials and implementation of novel electrode concepts resulting in:
  - High oxygen reduction reaction (ORR) activity, viable for practical automotive systems
  - Improved catalyst durability
  - High ionic/electronic conductivity within the catalyst layer
  - Adequate oxygen mass transport and effective removal of the water product

## Fiscal Year (FY) 2015 Objectives

• Optimize multiple nitrogen precursor synthesis approach for the development of advanced Fe-N-C catalysts

- Develop non-PGM cathode model based on microstructurally-consistent agglomerate parameters from nano-X-ray tomography (XRT) and electron microscopy
- Combine theoretical density functional theory (DFT) modeling with surface probe approaches, nuclear resonance vibrational spectroscopy (NRVS) and Δµ-X-ray absorption spectroscopy (XAS), for the identification of Fe-N<sub>x</sub> active sites
- Large-scale membrane electrode assembly (MEA) fabrication (50 cm<sup>2</sup>) in industry environment

#### **Technical Barriers**

This project addresses the following technical barriers from the Fuel Cells section (3.4.5) of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration (MYRDD) Plan [1]:

- (A) Durability (catalysts, electrode layers)
- (B) Cost (catalyst, MEAs)
- (C) Performance (catalysts, electrodes, MEAs)

#### **Technical Targets**

Non-PGM fuel cell cathode catalyst research in this project focuses on the DOE technical targets outlined in Table 3.4.13 in the Fuel Cells section (3.4.4) of the MYRDD Plan [1]. The ultimate technical targets of the project are:

- Catalyst activity in H<sub>2</sub>/O<sub>2</sub> MEA at 0.044 A cm<sup>-2</sup> (80°C): ≥0.87 V (*iR*-free)
- Four-electron selectivity (rotating ring-disk electrode [RRDE]): ≥99% (H<sub>2</sub>O<sub>2</sub> ≤1%)
- MEA maximum power density at  $80^{\circ}$ C:  $\geq 1.0$  W cm<sup>-2</sup>
- Performance loss at 0.80 A cm<sup>-2</sup> after 30,000 cycles in  $N_2$ :  $\leq$ 30 mV

**TABLE 1.** Progress towards Meeting Technical Targets for Electrocatalysts for

 Transportation Applications

Characteristic	Unit	Targets (CWG Proposed)		2015 Status
		2020	Ultimate	
Voltage at 0.044 A/cm <sup>2*</sup>	V <sub>iR-free</sub>	0.87	0.90	0.86

\* Test at 80°C H<sub>2</sub>/O<sub>2</sub> in MEA; fully humidified with total outlet pressure of 150 kPa (abs); anode stoichiometry 2; cathode stoichiometry 9.5 [2]. CWG – Catalysis Working Group

## FY 2015 Accomplishments

- An advanced cyanamide-polyaniline (CM-PANI)-Fe-C catalyst with favorable micro- and meso-structure was synthesized via the novel multiple nitrogen precursor approach. The catalyst was tested in H<sub>2</sub>/O<sub>2</sub> MEA (80°C) reaching a catalyst activity of 0.86 V (*iR*-free) at 0.044 A cm<sup>-2</sup>, only 0.01 V from the 2020 target (as proposed by DOE Catalysis Working Group).
- Promising initial activity was obtained from a Fe-free catalyst (CM-PANI-Co-C) in rotating disk electrode (RDE) (*E*<sup>1/2</sup> of 0.74 V) and fuel cell testing (with relatively low loading, 2.0 mg cm<sup>-2</sup>).
- Combination of DFT modeling, NRVS, and  $\Delta\mu$ -XAS studies points to the presence of surface active Fe-N<sub>x</sub> sites.
- A microstructurally consistent fuel cell model was validated against experimental H<sub>2</sub>-air fuel cell data with varying Nafion<sup>®</sup> loading.

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## INTRODUCTION

Cost studies estimate that Pt-based catalysts involve almost half of the entire polymer electrolyte fuel cell (PEFC) stack cost and as much as 20% of the overall system cost [3]. Since Pt is a precious metal, its cost will not benefit from economies of scale and is subject to volatile price fluctuations and monopolized global distributions. Reducing, or ideally replacing, expensive Pt and/or Pt-alloy catalysts in PEFC systems is highly desirable and has been a major focus of research and development efforts in fuel cell electrocatalysis. Owing to the inherently sluggish ORR occurring at the fuel cell cathode, higher Pt content is required at the cathode than at the anode. Successful development of non-PGM catalysts for ORR would provide the most significant economic advantage in PEFC systems. However, hindering the successful elimination of Pt cathode catalysts in PEFC systems is the lack of non-PGM catalysts that can provide sufficiently high ORR activity and, especially, the durability under the conditions of fuel cell cathode operation.

## **APPROACH**

In this research project, we intend to accomplish major advancements in non-PGM cathode technology through the development and implementation of novel materials and concepts. Our catalyst development effort has concentrated on novel synthesis methods, including high temperature catalyst synthesis using multiple nitrogen-containing precursors, advanced carbon supports, and in an earlier stage of the project, non-pyrolyzed phthalocyanine-derived catalysts, as well as metal-free catalysts based on nitrogendoped carbon nanostructures. Comprehensive testing of materials, including initial performance screening by in situ electrochemical techniques and ex situ characterization to assess catalyst activity and durability, identify catalytic sites and validate fuel cell performance of the most promising materials, represents a substantial fraction of the efforts.

The use of non-PGM ORR catalysts results in cathodes with increased thickness compared to that of Pt-based cathodes. Thus, significant effort is required to address the resulting electrode design challenges. Key issues include oxygen mass transport, proton conductivity and prevention of catalyst layer flooding. Our research has focused on the validation of existing General Motors electrode model for non-PGM electrodes and parameter approximation using in situ microstructured electrode scaffold (MES) diagnostics. Electrode optimization is based on the insight obtained from the modeling, nanoscale XRT imaging, and advanced microscopy analysis. In parallel to the catalyst and electrode development components of this project, MEA fabrication, optimization and scale-up are being performed to obtain a  $50 \text{ cm}^2$  (or larger, if needed) MEA with the best performing materials for independent testing and evaluation at a DOEapproved facility.

## RESULTS

Achieved fuel cell voltage of 0.86 V (iR-free) at  $0.044 \text{ A cm}^{-2}$  in  $H_{\gamma}/O_{\gamma}$  fuel cell testing. An advanced non-PGM catalyst developed according to LANL's dual nitrogen precursor approach was synthesized, optimized, and tested for fuel cell performance. A fuel cell operating with advanced CM-PANI-Fe-C catalyst achieved a voltage of 0.86 V (*iR*-free) at 0.044 A cm<sup>-2</sup> in  $H_2/O_2$  MEA testing at 80°C (Figure 1a). This fuel cell voltage is only 0.01 V below the 2020 target voltage of 0.87 V (see Table 1 above). Further, the advanced CM-PANI-Fe-C catalyst demonstrated superior durability with potential loss at  $E_{\nu_a} \sim 30$  mV after 30,000 cycles between 0.2 V and 1.0 V in N<sub>2</sub>-saturated electrolyte. This durability was accompanied by less than 2% H<sub>2</sub>O<sub>2</sub> yield in the entire ORR potential range (Figure 1b). These results achieve and exceed the June 2015 durability and selectivity targets.

DFT modeling of most active reported structure to date,  $FeCoN_5(OH)$ , based on thermodynamic stability arguments. Calculation of active site structures was performed using thermodynamic limiting potential,  $U_L$ , with a computational hydrogen electrode and DFT of ORR intermediate binding energies. In the developed model, \*OH ligand spontaneously formed in water-bearing environments, modifying the active site structure and thus improving predicted ORR activity. Higher limiting potentials were obtained from bi-metallic sites.  $U_L$  for Fe<sub>2</sub>N<sub>5</sub>(OH) was calculated to be 0.72 V while a mixed-metal active site structure, FeCoN<sub>5</sub>(OH), resulted in



**FIGURE 1.** (a) Fuel cell performance of advanced CM-PANI-Fe-C non-PGM catalyst demonstrating fuel cell voltage of 0.86 V (*iR*-free) at 0.044 A cm<sup>-2</sup> in H<sub>2</sub>-O<sub>2</sub>. Inset: Pore size distribution data for catalyst tested. Anode: 0.2 mg<sub>Pt</sub> cm<sup>-2</sup> Pt/C H<sub>2</sub>, 50 sccm, 1.0 bar H<sub>2</sub> partial pressure; cathode: ~4.0 mg cm<sup>-2</sup> air, 100 sccm, 1.0 bar air partial pressure; membrane: Nafion<sup>®</sup> 117; cell size: 5 cm<sup>2</sup>. (b) RDE durability cycling of advanced CM-PANI-Fe-C catalyst demonstrating potential loss ~30 mV  $E_{\frac{1}{2}}$  after 30,000 cycles in N<sub>2</sub>-saturated electrolyte. Inset: cyclic voltammogram showing no surface area change after durability cycling. RRDE: 0.5 M H<sub>2</sub>SO<sub>4</sub>; 900 rpm; 25°C; Hg/HgSO<sub>4</sub> (0.5 M H<sub>2</sub>SO<sub>4</sub>) reference electrode; graphite counter electrode; steady-state potential program: 30 mV steps, 30 s/step; cyclic voltammetry: 0.5 M H<sub>2</sub>SO<sub>4</sub>; 0 rpm; 25°C; N<sub>2</sub>-saturated solution; 20 mV/s.

the most active structure reported to date from computational studies: 0.80 V.

Validation of surface-probe XAS approach with theoretical modeling for identification of active site structures. In situ surface-sensitive XAS/ $\Delta\mu$ -X-ray absorption near edge spectroscopy (XANES) analytical technique was utilized to monitor changes in Fe-N<sub>x</sub>-O species while under fuel cell and RDE operation. Experimental  $\Delta\mu$  results obtained are in good agreement with theoretical DFTderived model structures: Fe<sub>2</sub>N<sub>6</sub>, FeN<sub>4</sub>, and FeN<sub>5</sub> (Figure 2a). Monitoring of  $\Delta\mu$  changes after potential cycling under inert  $(N_2)$  and reaction (air) conditions indicates that Fe-N<sub>x</sub> centers are likely responsible for ORR activity (Figure 2b).

Separate mapping of catalyst and Nafion<sup>®</sup> distribution and validation of fuel cell cathode model. The advanced non-PGM catalysts prepared from multiple nitrogen precursors were integrated into electrodes containing Nafion<sup>®</sup> ionomer and studied using nano-XRT for the development of microstructurally-consistent cathode model (Figure 3a). Staining of Nafion<sup>®</sup> with Cs<sup>+</sup> allowed for the identification of the distribution of the ionomer layer in thick cathodes to separately map catalyst (Zernike phase contrast) and Nafion<sup>®</sup>



**FIGURE 2.** (a) Experimental  $\Delta\mu$  data showing good agreement with theoretical DFT-derived Fe-N<sub>x</sub> structures: FeN<sub>5</sub>, FeN<sub>4</sub>, Fe<sub>2</sub>N<sub>6</sub>. (b) Experimental  $\Delta\mu$  data as a function of cycling suggesting Fe-N<sub>y</sub> centers are likely responsible for ORR activity. Cycling: 3,000 cycles, H<sub>2</sub>/N<sub>2</sub> and H<sub>2</sub>/air.



**FIGURE 3.** (a) Microstructurally consistent model framework showing the process involved in the development of a cathode model based on morphological property distribution inputs and transport property simulations. (b) 3D Nafion<sup>®</sup> density maps obtained from Cs<sup>+</sup> staining of Nafion<sup>®</sup> for two different loadings, 35 wt% and 60 wt%. (c) Simulated and experimental  $H_2$ -air fuel cell polarization plots validating cathode model obtained from morphological parameters observed and calculated from nano-XRT for various Nafion<sup>®</sup> loadings.

(absorption contrast) at up to 50 nm resolution (Figure 3b). At a lower Nafion<sup>®</sup> loading (35 wt%), the ionomer forms clumps with low infiltration into the dense pores of the catalyst. While at higher Nafion<sup>®</sup> loadings (50–60 wt%), infiltrated ionomer forms thick films on the surface.

A cathode model was developed based on microstructurally consistent morphological properties obtained from nano-XRT imaging along with transport property simulations. This model was validated against  $H_2$ -air fuel cell experiments at varying Nafion<sup>®</sup> loading (Figure 3c). A key finding of the two-phase model is a notable performance improvement obtained by mitigation of liquid water flooding of thicker electrodes. Results suggest that carbon agglomerate size distribution has a significant impact on fuel cell performance, proving predictions from single diameter model insufficient. The combination of mapping of catalyst-Nafion<sup>®</sup> distribution along with fuel cell performance at varying ionomer loading suggests that an optimal dispersion of ionomer within carbon-rich catalyst in the cathode is an essential strategy for increasing catalyst utilization, thereby significantly improving catalyst performance.

Microstructured Electrode Scaffold (MES) for direct ORR and conductivity measurements. Figure 4a displays the MES schematic used for direct conductivity measurement during PEFC operation. The local ORR rate and potential distribution of Nafion<sup>®</sup> were measured as a function of distance from the membrane for three different relative humidity (RH) conditions: 50%, 80%, 140%. An RH of 50% showed limited proton transport with the ORR primarily occurring near the proton exchange membrane. On the other hand, 140% RH showed increased proton conductivity as a result of water production. Further, a large *iR*-drop due to water accumulation gave rise to the second ohmic slope, evident in catalyst-coated membrane (CCM) studies with high Nafion<sup>®</sup> loading and consistent with modeling studies. ORR at 140% RH occurs mainly near the gas diffusion layer (Figure 4b).



**FIGURE 4.** (a) Schematic of MES used for direct conductivity measurement during PEFC operation. (b) Potential distribution of Nafion<sup>®</sup> as a function of distance from membrane for three different RH conditions: 50%, 80%, 140%. An RH of 50% shows limited proton transport, while 140% RH shows increased proton conductivity as a result of water production. The large *iR*-drop observed for 140% RH is due to water accumulation giving rise to the second Ohmic slope and validating the model.

Electron microscopy analysis of state-of-the-art LANL catalysts and electrodes. Advanced scanning electron microscopy, transmission electron microscopy, and scanning transmission electron microscopy were performed to study the advanced CM-PANI-Fe-C catalyst material in order to provide insight into the structure of high-performance catalysts with favorable micro- and meso-porosity. Figure 5 shows the CM-PANI-Fe-C morphology consisting of fibrous carbonaceous agglomerates (randomly oriented, intertwined graphitic domains) and layered graphene sheets. Presence of graphene sheets are mostly associated with Fe/FeS particles. Annular dark-field-STEM imaging in combination with high resolution electron energy loss spectroscopy analysis was used to identify the presence of Fe atoms dispersed on the surface of few-layer graphene sheets. These Fe atoms were highly mobile under the electron beam.

## CONCLUSIONS

- Continued development of the high surface area multiple nitrogen precursor CM-PANI-Fe-C catalyst led to notable improvement in H<sub>2</sub>-air performance resulting in a fuel cell voltage of 0.86 V obtained at 0.044 A cm<sup>-2</sup> (nearing the proposed 2020 target of 0.87 V).
- ORR selectivity and durability targets for June 2015 were achieved and exceeded with advanced CM-PANI-Fe-C catalyst:
  - Potential loss of ~30 mV at  $E_{\frac{1}{2}}$  after 30,000 cycles between 0.2 V and 1.0 V.
  - $H_2O_2$  yield of less than 2% across the entire potential range.

- Theoretical DFT modeling studies found that multi-metal  $Fe_2N_5$  sites are likely the key moiety, in agreement with NRVS-NO<sub>(0)</sub> and XAS/ $\Delta\mu$ -XANES probing.
- A cathode model was developed using morphological property distribution inputs from nano-XRT imaging and transport property simulations. The model was validated against experimental  $H_2$ -air fuel cell studies with different Nafion<sup>®</sup> loadings.

## **FUTURE DIRECTIONS**

- Catalyst Development:
  - Completion of remaining activity/durability catalyst performance targets
  - Enhancement of the ORR activity of Fe-free catalysts
  - Further development of two-nitrogen precursor catalysts, focusing on activity and durability
- Active Site and Durability Studies:
  - In situ XAS studies in combination with DFT-FEFF calculations for active site determination
  - Completion of the Mössbauer study of active sites using molecular probes
  - Determination of corrosion rate measurements; comparison with Pt/C catalysts
- Electrode Design and Modeling:



**FIGURE 5.** Microscopy studies for advanced CM-PANI-Fe-C catalyst showing two morphological features. The primary feature consists of fibrous carbonaceous agglomerates which are randomly oriented and have an intertwined graphitic domain. The secondary morphological feature consists of few-layer graphene sheets with associated Fe/FeS particles and Fe atoms dispersed atop graphene.

- Electrode structure and modeling analysis of scaled IRD and General Motors electrodes using nano-XRT
- Detailed modeling of MES system for model improvement; parametric studies of electrode formulation by MES
- MEA Optimization and Fabrication:
  - Completion of electrode optimization study
  - Optimization of the first generation spray-coated MEAs
  - Fabrication and testing of 50 cm<sup>2</sup> MEAs

#### SPECIAL RECOGNITIONS & AWARDS/ PATENTS ISSUED

**1.** Piotr Zelenay awarded Los Alamos National Laboratory Fellows Prize "for longstanding contributions to the understanding of non-precious metal electrocatalysts for fuel cells," February 2015.

**2.** Piotr Zelenay awarded Fellowship of the Electrochemical Society "for major contributions in the development of materials and concepts for polymer electrolyte fuel cells," October 2014.

## **FY 2015 PUBLICATIONS**

**1.** H.T. Chung and P. Zelenay, "A Simple Synthesis of Nitrogen-Doped Carbon Micro- and Nanotubes," *Chem. Comm.*, DOI: 10.1039/C5CC04621A (2015).

**2.** Q. Jia, N. Ramaswamy, H. Hafiz, U. Tylus, K. Strickland, G. Wu, B. Barbiellini, A. Bansil, E.F. Holby, P. Zelenay, and S. Mukerjee, "Dynamic Nature of Active Sites in Iron-based Catalysts during Electrocatalysis," *Nat. Chem.* submitted.

**3.** U. Martinez, G.M. Purdy, E.F. Holby, K. Artyushkova, J.H. Dumont, A. Singh, N.H. Mack, P. Atanassov, D.A. Cullen, K.L. Moore, M. Chhowalla, P. Zelenay, A.M. Dattelbaum, A.D. Mohite, and G. Gupta, "Critical Role of the Removal of Intercalated Water for Electrocatalytically Active Graphitic Systems," *Nat. Chem.*, submitted.

**4.** L. Chen, G. Wu, E.F. Holby, P. Zelenay, W. Tao, and Q. Kang, "Lattice Boltzmann Investigation of Reactive Transport in Cathode Catalyst Layers of Proton Exchange Membrane Fuel Cells," *Electrochim. Acta*, **158**, 175–186 (2015).

**5.** D. Higgins, F. Hassan, M. Seo, J. Choi, A. Hoque, D. Lee, and Z. Chen, "Shape-Controlled Octahedral Cobalt Disulfide Nanoparticles Supported on Nitrogen and Sulfur-Doped Graphene/ Carbon Nanotube Composites for Oxygen Reduction in Acidic Electrolyte," *J. Mat. Chem. A* **3**, 6340–6350 (2015). **6.** E.F. Holby, G. Wu, P. Zelenay, and C.D. Taylor, "Structure of Fe-N<sub>x</sub>-C defects in Oxygen Reduction Reaction Catalysts from First Principles Modeling," *J. Phys. Chem. C*, **118** (26) 14388–14393 (2014).

**7.** S. Komini Babu, H. Chung, G. Wu, P. Zelenay, and S. Litster, "Modeling Hierarchical Non-Precious Metal Catalyst Cathodes for PEFCs Using Multi-Scale X-ray CT Imaging," *ECS Trans.*, **64** (3), 281–192 (2014).

**8.** U. Martinez, T.L. Williamson, K. Artyushkova, N.H. Mack, G.M. Purdy, J.H. Dumont, D. Kelly, W. Gao, A. M. Dattelbaum, A. Mohite, G. Gupta, and P. Zelenay, "Thin-Film Non-Precious Metal Model Catalysts for Oxygen Reduction Reaction," *ECS Trans.*, **64** (3), 293–301 (2014).

**9.** Q. Li, G. Wu, N.H. Mack, H.T. Chung, P. Xu, H.-L. Wang, S. Ma, and P. Zelenay, "Phosphate-Tolerant Oxygen Reduction Catalysts," *ACS Catal.*, **4** (9), 3193–3200 (2014).

**10.** P. Zelenay, H.T. Chung, E.F. Holby, C.D. Taylor, and G. Wu, "Carbon Composite Non-Precious Metal Catalysts for Oxygen Reduction in Electrochemical Energy Conversion," *Prepr. Pap.-Am. Chem. Soc., Div. Fuel Chem.*, accepted (2014).

**11.** R. Mukundan, D. Spernjak, G. Wu, D. Hussey, D. Jacobson, A. Steinbach, R. Borup, and P. Zelenay, "Visualizing Water in Non-Precious Metal Catalyst-based Polymer Electrolyte Fuel Cells Using Neutron Imaging," *Prepr. Pap.-Am. Chem. Soc., Div. Energy Fuels*, accepted, (2014).

**12.** E.T. Holby, G. Wu, P. Zelenay, and C.D. Taylor, "Structure of Fe-N<sub>x</sub>-C Defects in Oxygen Reduction Reaction Catalysts from First Principles Modeling," *J. Phys. Chem. C*, **118**, 14388–14393 (2014).

**13.** H.T. Chung, G. Wu, Q. Li, and P. Zelenay, "Role of Two Carbon Phases in Oxygen Reduction Reaction on the Co-PPy-C Catalyst," *Int. J. Hydrogen Energy*, **39**, 15887–15893 (2014).

**14.** D.C. Higgins, G. Wu, H.T Chung, U. Martinez, S. Ma, Z. Chen, and P. Zelenay, "Manganese-Based Non-Precious Metal Catalysts for Oxygen Reduction in Acidic Media," *ECS Trans.*, **61** (31) 35–42 (2014).

**15.** M. Seo, D. Higgins, G. Jiang, S. Choi, B. Han, and Z. Chen, "Theoretical Insight into Highly Durable Iron Phthalocyanine Derived Non-Precious Catalysts for Oxygen Reduction Reactions," *J. Mat. Chem. A* **2**, 19707–19616 (2014).

**16.** P. Zamani, D. Higgins, F. Hassan, G. Jiang, J. Wu, S. Abureden, and Z. Chen, "Electrospun Iron–Polyaniline–Polyacrylonitrile Derived Nanofibers as Non–Precious Oxygen Reduction Reaction Catalysts for PEM Fuel Cells," *Electrochim. Acta*, **139**, 111–116 (2014).

## **FY 2015 PRESENTATIONS**

**1.** S. Litster, "Fuel cell and battery electrodes: Advanced characterization and new electrode concepts," University of Victoria, Victoria, Canada, June 29 (2015) – *invited lecture*.

**2.** S. Litster, "Nano-scale X-ray computed tomography applied to fuel cell and battery electrode characterization and optimization," Naval Research Laboratory, Washington, DC, June 11 (2015) – *invited lecture*.

**3.** D. Higgins, H. Chung, U. Tylus, Z. Chen, and P. Zelenay, "Combined Nitrogen Precursor Approach to Develop Cobalt-Based Non-Precious Catalysts for Polymer Electrolyte Fuel Cell Cathodes," 227th Meeting of the Electrochemical Society, Chicago, Illinois, USA, May 24–28 (2015).

**4.** J.H. Dumont, U. Martinez, A. Mohite, G.M. Purdy, A.M. Dattelbaum, P. Atanassov, and P. Zelenay, G. Gupta, "Graphene Oxide Based Non Precious Metal Catalysts for Oxygen Reduction Reaction in Alkaline Media," 227th Meeting of the Electrochemical Society, Chicago, Illinois, USA, May 24–28 (2015).

5. H. Chung, D. Higgins, D. Kim, G. Wu, U. Tylus, K. More, D. Cullen, P. Zelenay, "An Approach for Highly Porous Non-Precious Metal Catalyst Synthesis for Polymer Electrolyte Fuel Cell Cathodes," 227th Meeting of the Electrochemical Society, Chicago, Illinois, USA, May 24–28 (2015).

**6.** U. Martinez, T. Williamson, K. Artyushkova, M. Hoffbauer, G. Purdy, J. Dumont, A. Dattelbaum, A. Mohite, G. Gupta, and P. Zelenay, "Model Non-Precious Metal Catalysts for Oxygen Reduction Reaction: A Bottom-up Approach," 227th Meeting of the Electrochemical Society, Chicago, Illinois, USA, May 24–28 (2015).

**7.** U. Tylus, H. Chung, D. Higgins, D. Myers, D. Nordlund, C. Segre, and P. Zelenay, "Study of Non-PGM ORR Catalyst Degradation Using Synchrotron Techniques," 227th Meeting of the Electrochemical Society, Chicago, Illinois, USA, May 24–28 (2015).

**8.** S. Litster, "Nano-scale X-ray computed tomography applied to fuel cell and battery electrode characterization and optimization," National Institute of Advanced Industrial Science and Technology (AIST) Kansai, Osaka, Japan, May 12 (2015) – *invited lecture*.

**9.** S. Litster, "Nano-scale X-ray computed tomography applied to fuel cell and battery electrode characterization and optimization," Chemical Engineering, Kyoto University, Kyoto, Japan, May 11 (2015) – *invited lecture*.

**10.** H. Chung, E. Holby, U. Martinez, G. Purdy, J. Dumont, U. Tylus, D. Higgins, G. Gupta, A. Dattelbaum, Z. Chen, and P. Zelenay, "Oxygen Reduction Electrocatalysts for Fuel Cell Applications," Materials Research Society Meeting & Exhibit, San Francisco, California, April 6–10 (2015) – *invited lecture*.

**11**. P. Zelenay, "A Few Highlights from Fuel Cell Electrocatalysis Research at Los Alamos," CARISMA 2014 - International Fuel Cell Conference, Cape Town, South Africa, December 1–3 (2014) – *invited lecture*.

**12.** G. Wu, H.T. Chung, E.F. Holby, and P. Zelenay, "Oxygen Reduction on Non-Precious Metal Fuel Cell Catalysts," Henan Normal University, Xinxiang, China, November 5 (2014) – *invited plenary lecture*.

**13.** P. Zelenay, "Electrocatalysts for Fuel Cell Applications," International Conference on Energy Science and Technology (EEST2014), Shanghai, China, November 1–3 (2014) – *invited plenary lecture*.

**14.** E.F. Holby, C.D. Taylor, and P. Zelenay, "Modeling Non-Precious Metal Catalyst Structures for Oxygen Reduction," International Symposium on Electrocatalysis: Explorations of the Volcano Landscape, Whistler, British Columbia, Canada, October 26–29 (2014).

**15.** P. Zelenay, "Oxygen Reduction on Non-Precious Metal Electrocatalysts: Accomplishments and Challenges," International Symposium on Electrocatalysis: Explorations of the Volcano Landscape, Whistler, British Columbia, Canada, October 26–29 (2014) – *invited keynote lecture*.

**16.** U. Martinez, E.F. Holby, G.M. Purdy, G. Gupta, A. Mohite, G. Wu, H.T. Chung, A.M. Dattelbaum, and P. Zelenay, "Model Systems and Modeling of Non-precious Metal Oxygen Reduction Catalysts," 226th Meeting of the Electrochemical Society, Cancun, Mexico, October 5–10 (2014) – *invited lecture*.

**17.** U. Martinez, T.L. Williamson, G.M. Purdy, J.H. Dumont, N.H. Mack, A.M. Dattelbaum, A. Mohite, G. Gupta, and P. Zelenay, "Thin-Film Non-Precious Metal Model Catalysts for Oxygen Reduction Reaction," 226th Meeting of the Electrochemical Society, Cancun, Mexico, October 5–10 (2014).

**18.** S.K. Babu, H.T. Chung, G. Wu, P. Zelenay, and S. Litster, "Modeling Hierarchical Non-Precious Metal Catalyst Cathodes for PEFCs using Multi-Scale X-ray CT Imaging Data," 226th Meeting of the Electrochemical Society, Cancun, Mexico, October 5–10 (2014).

**19.** U. Martinez, A. Mohite, G. M. Purdy, A. M. Dattelbaum, P. Zelenay, and G. Gupta, "Highly Tunable and Ordered Graphene-Oxide-Based Materials for Energy Applications," 226th Meeting of the Electrochemical Society, Cancun, Mexico, October 5–10 (2014).

**20.** P. Zelenay, "Non-Precious Metal Electrocatalysts: Accomplishments and Challenges," "226th Meeting of the Electrochemical Society, Cancun, Mexico, October 5–10 (2014) – *invited lecture*.

**21.** R. Borup, R. Mukundan, D. Spernjak, G. Wu, A. Steinbach, P. Zelenay, D. Hussey, and D. Jacobson, "Neutron Imaging to Optimize PEM Fuel Cells Performance," 10th World Conference on Neutron Radiography (WCNR-10), Grindelwald, Switzerland, October 5–10 (2014).

**22**. P. Zelenay, "Oxygen Reduction on Non-Precious Metal Fuel Cell Electrocatalysts," Department of Chemistry, University of Indiana, Bloomington, Indiana, September 23 (2014) – *invited lecture*.

**23.** P. Zelenay, H.T. Chung, E.F. Holby, C.D. Taylor, and G. Wu, "Carbon Composite Non-Precious Metal Catalysts for Oxygen Reduction in Electrochemical Energy Conversion," 248th American Chemical Society Meeting and Exposition, San Francisco, California, August 10–14 (2014).

**24.** R. Mukundan, D. Spernjak, G. Wu, D. Hussey, D. Jacobson, A. Steinbach, R. Borup and P. Zelenay, "Visualizing Water in Non-Precious Metal Catalyst-based Polymer Electrolyte Fuel Cells Using Neutron Imaging," 248th American Chemical Society Meeting and Exposition, San Francisco, California, August 10–14 (2014). (invited lecture).

**25.** S. Komini Babu, H.T. Chung, G. Wu, P. Zelenay, and S. Litster, "Modeling Hierarchical Non-Precious Metal Catalyst Cathodes for PEFCs using Multi-Scale X-Ray CT Imaging," Gordon Research Conference on Fuel Cells, Smithfield, Rhode Island, August 3–8 (2014) –*received the best student poster award*.

**26.** P. Zelenay, "A Few Highlights from Fuel Cell Electrocatalysis Research at Los Alamos," International Symposium on Clean Energy from Ethanol, ISCEE 2014, Rzeszow, Poland, July 28–31 (2014) – *invited keynote lecture*.

**27.** P. Zelenay, "Modern Fuel Cell Electrocatalysis," AGH University of Technology, Krakow, Poland, July 23 (2014) – *invited lecture*.

**28.** P. Zelenay, "Electrocatalysis in Fuel Cells," Industrial Chemistry Research Institute, Warsaw, Poland, July 22 (2014) – *invited lecture*.

#### REFERENCES

**1.** *Multi-Year Research, Development and Demonstration Plan: Section 3.4 Fuel Cells*, Fuel Cell Technologies Office, 2011. http://www1.eere.energy.gov/hydrogenandfuelcells/mypp/pdfs/fuel\_cells.pdf

2. H. Gasteiger et al., Appl. Catal. B-Environ., 56, 9-35, 2005.

**3.** B. James et al., DTI, Inc., 2010 DOE Hydrogen Program Review, Washington, DC, June 9, 2010.