DOE Fuel Cells and Infrastructure Technologies 2009 Kickoff Meeting

The Science And Engineering of Durable Ultralow PGM Catalysts

Los Alamos National Laboratory (lead participant)
Ballard Power Systems (BPS)
University of California Riverside (UCR)
University of New Mexico (UNM)
Oak Ridge National Laboratory (ORNL)

Eric Brosha, Neil Henson and Fernando Garzon (PI) LANL

Los Alamos NATIONAL LABORATORY

The Institute for Hydrogen and Fuel Cell Research

Project Objectives

- *Development of durable, high mass activity Platinum Group Metal cathode catalysts*

- *Elucidation of the fundamental relationships between PGM catalyst shape, particle size and activity*

- *Optimization of the cathode electrode layer to maximize the performance of PGM catalysts*

- *Understanding the performance degradation mechanisms of high mass activity cathode catalysts*

- *Development and testing of fuel cells using ultra-low loading high activity PGM catalysts*

Technical Targets/Barriers

Technical Targets:
Decrease PGM content while increasing mass activity and lifetime

Table 3.4.12 Technical Targets: Electrocatalysts for Transportation Applications

Characteristic	Units	2005 Status [a]		Stack Targets	
		Cell	Stack	2010	2015
Platinum group metal total content (both electrodes)	g / kW (rated)	0.6	1.1	0.3	0.2
Platinum group metal (pgm) total loading [b]	mg PGM / cm^2 electrode area	0.45	0.8	0.3	0.2
Cost	$ / kW	9	55 [c]	5 [d]	3 [d]
Durability with cycling					
Operating temp ≤80°C	hours	>2,000	~2,000 [e]	5,000 [f]	5,000 [f]
Operating temp >80°C	hours	N/A [g]	N/A [g]	2,000	5,000 [f]
Electrochemical area loss [h]	%	90	90	<40	<40
Electrocatalyst support loss [h]	mV after 100 hours @ 1.2V	>30 [i]	N/A	<30	<30
Mass activity [j]	A / mg Pt @ 900 $mV_{iR\text{-}free}$	0.28	0.11	0.44	0.44
Specific activity [j]	μA / cm^2 @ 900 $mV_{iR\text{-}free}$	550	180	720	720
Non-Pt catalyst activity per volume of supported catalyst	A / cm^3 @ 800 $mV_{iR\text{-}free}$	8	N/A	>130	300

Barriers:

- PGM catalysts are difficult to synthesize in configurations other than quasi-spherical particles
- PGM area specific activity may decrease with decreasing particle size
- Durability may decrease with greater PGM surface area to volume ratios

Approach

- *Use contemporary theoretical modeling and advanced computational methods to understand and engineer the new catalysts*

- *Model and design appropriate catalyst architectures to maximize the performance of our novel catalysts*

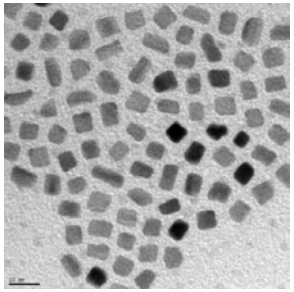
- *Investigate catalyst-support interactions and their effects on durability and mass activity will also be investigated*

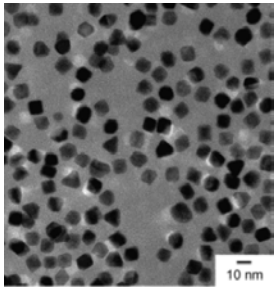
- *Study and test the performance of the catalysts in electrochemical cells, single cell-fuel cells and fuel cell stacks*

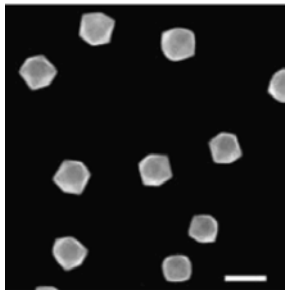
- *Extensively characterize new materials before and after fuel cell operation*

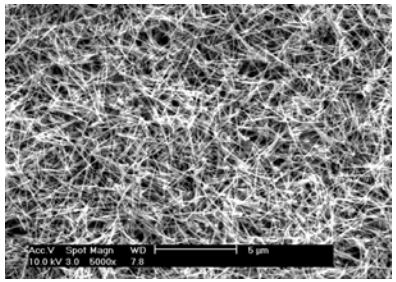
Approach

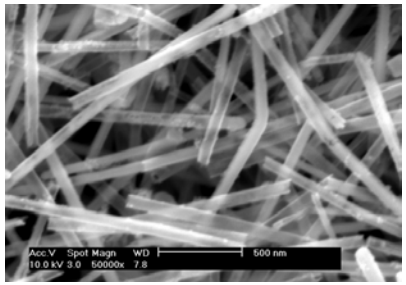
- Synthesis of new PGM materials differing shapes and sizes: cubes, octahedron, tetrahexahedral, wires and tubes etc.
- TEM SEM and X-ray, neutron characterization of structures
- Electrochemical ORR kinetics
- Electrochemical and calorimetric studies of stability
- Optimization of electrode structure
- Fuel cell testing
- DFT modeling of catalyst activity and particle stability

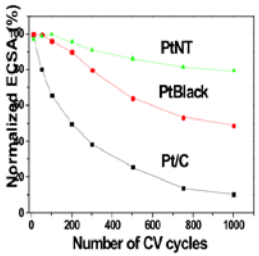


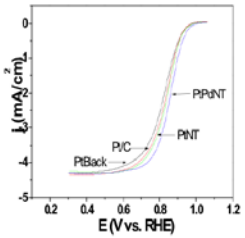


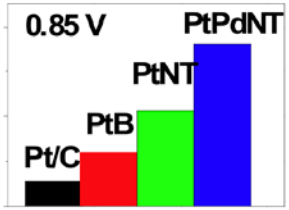








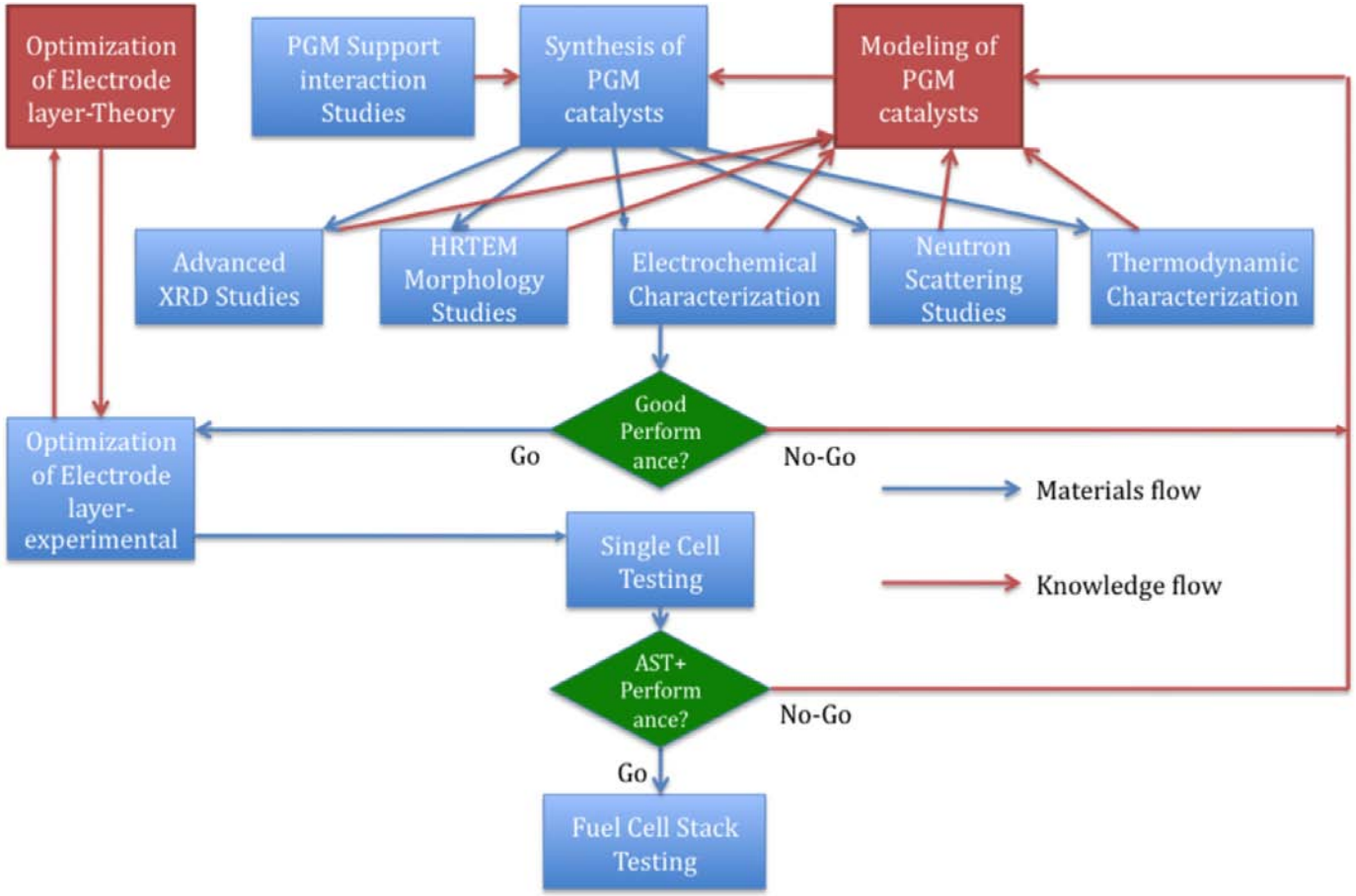




Task Assignments

- Theoretical Understanding Of Roles Of PGM Catalyst Shape, Size, Support Interactions And Catalyst Layer Architecture On Cathode Mass Activity And Durability
  - Optimization Of PGM Catalyst Morphology With Guidance From Computational Studies (LANL)
  - Optimization Of Catalyst Layer Architecture With Guidance From Microstructural Simulations (Ballard)
- Experimental Synthesis And Characterization Of New Geometry PGM Catalysts
  - Synthesis of Novel Pt Nanoparticles (UCR, UNM, LANL)
  - Synthesis of Pt Nanotubes (UCR)
  - Synthesis of Pt Nanowires (UCR UNM)
- PGM Structural Characterization by TEM, XRD, Neutron Scattering
  - HRTEM Morphology Studies (ORNL)
  - Advanced X-ray Diffraction Studies (LANL)
  - Neutron Scattering Studies (LANL)
  - Thermodynamic Characterization of PGM catalysts (UNM LANL)
  - Electrochemical Characterization of PGM catalysts (LANL)
- Understanding Catalyst Nucleation And Support Interactions
  - Inverse Chromatography Studies Of Precursor-Support Interactions (LANL UNM)
  - PGM-Support Interaction Studies (UNM)
- Fuel Cell Testing Of Novel PGM Catalysts (LANL Ballard)
  - Testing of novel catalysts in fuel cells (LANL Ballard)
  - Fuel cell post testing materials characterization (LANL ORNL)

Technical Approach



Organization/Participants

National Labs

- LANL
  - *Fernando Garzon*
  - *Eric Brosha*
  - *Mahlon Wilson*
  - *Neil Henson*

Universities

- UNM
  - *Prof. Abhaya Datye*
- UCR
  - *Prof. Yushan Yan*

Industry

- Ballard
  - *Siyu Ye*
  - *David Harvey*

Timeline

Project initiated in Sept FY2009 for 4 years

VASP/Gaussian modeling of PGMs		Stability and support-interaction modeling	
2010	2011	2012	2013

Modeling and Optimization of catalyst layer architectures

Synthesis and characterization of novel PGM's and supports

ORR studies

Fuel Cell performance testing

Catalyst/support interaction studies

Calorimetric investigations of PGM materials

Tasks, Milestones and Decision Points

Milestones and Decision Points

Task ID	Task Title	Quarter Due	Milestone Description
1.1	Modeling	3	VASP Modeling pf PGM catalyst
		4	GAUSSIAN03/MondoSCF modeling of PGM clusters
		8	Modeling of nano-particle reactivity
		12	Electrode-support calculations
1.2	Optimization of catalyst Layer Architecture	4	Computer generation of microstructures
		1-16	Visual optimization
		4,8,12	Process decision(s) – guidance and feedback, report.
		6	Go/No-Go
2.1	Synthesis of Novel Pt Nano-particles	4,8,12	Preparation of novel Pt nano-scale catalysts - UCR
		4,8,12	Preparation of novel Pt nano-scale catalysts - UNM
		4, 8, 12	Preparation of novel Pt nano-scale catalysts - Brosha
		8, 12	Process decision on synthesis, report.
2.2	Synthesis of Pt Nano-tubes	10	Go/No Go
2.3	Synthesis of Pt nanowires	12	Go/No Go
3.1	HRTEM Morphology Studies	4,8,12	TEM Investigation of nanoparticles, tubes, wires, and catalyst layer formations, report.
		6,10,14	Go/No Go decision on preparation
3.2	X-ray Diffraction Studies	4,8,12	XRD Investigation of nanoparticles, tubes, wires, and catalyst layer formations, report.
		6,10,14	Go/No Go decision on preparation
3.3	Neutron Scattering Experiments	8, 12	Experimental
		12, 14	PDF analysis
3.4	Thermodynamic Characterization of PGM Catalysts	4	Specific heat measurements
		8	Heat of sintering determination
		12	Assessment and theory
3.5	Electrochemical Characterization	8	RDE of nanoparticles
		10	RDE of nanotubes
		10	RDE of nanowires
		10	Go/No Go RDE v. CV
4.1	Precursor Support interaction studies	12	Determine precursor salt/support interaction parameters
4.2	PGM – Support interaction studies	12	Rates of emissions of adatoms from nanoparticles
		14	Modeling of loss mechanism
5.1	Fuel cell testing – single cell	8	Optimize MEA
		12	Evaluate catalysts surface area, particle size changes
		14	Go/No go
5.2	Durable electrode structure	12	MEA Prep
		14	Go/No go
5.3	Advanced Fuel cell testing	12	50 cm^2 testing
		14	Go/No go
		16	Deliver single cells and short stacks to designated DOE

Task Schedule, by Quarters (Q) with Milestones (M) and Decision Points (G)

Q / Task	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
1.1			M	M				M				M				
1.2				M		G		M				M				
2.1				M M M				M M M				M M M				
2.2										G						
2.3												G				
3.1				M		G		M		G		M		G		
3.2				M		G		M		G		M		M G		
3.3								M				M M		M		
3.4				M				M				M				
3.5								M		M M M G						
4.1												M				
4.2												M				
5.1								M				M		M G		
5.2												M		G		
5.3												M		G		D

Los Alamos NATIONAL LABORATORY

The Institute for Hydrogen and Fuel Cell Research

Budget

DOE Cost Share	Recipient Cost Share	Total
6,000,000	528,685	6,528,685
92%	8%	100

Yr 1	Yr 2	Yr 3	Yr 4	Cumulative
1,500,000	1,550,000	1,475,000	1,475,000	6,000,000

Participant	FY09-10 (Year 1)
LANL	$1,400,000
ORNL	$100,000
Universities	$275,000
Industry (Ballard)	$158,000
TOTAL Year 1	$1,933,000