U.S. DEPARTMENT OF ENERGY | Office of Science

Fundamental Research Related to Hydrogen and Fuel Cells

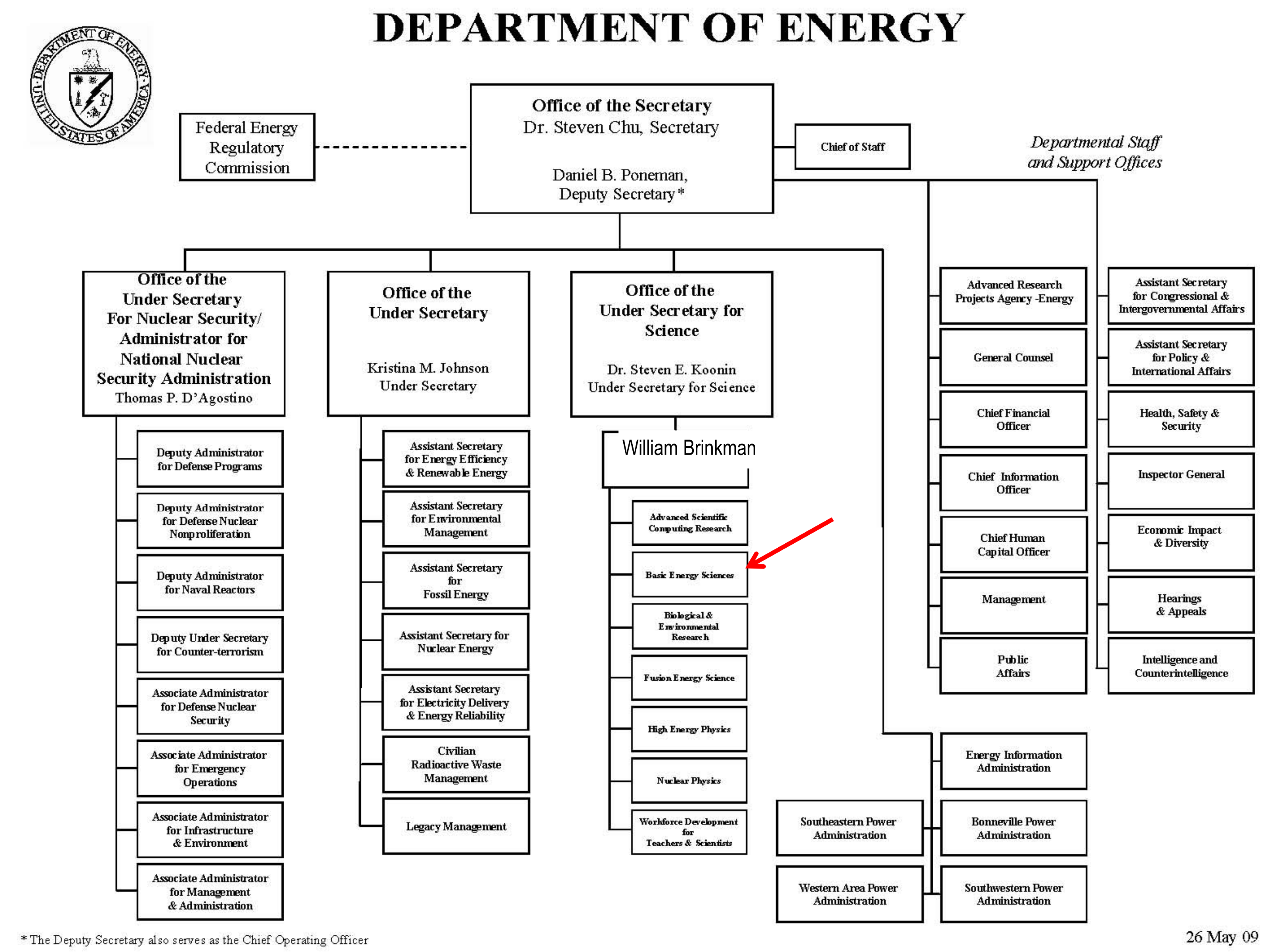
John Vetrano

Office of Basic Energy Sciences

Hydrogen Technical Advisory Committee Meeting

February 23, 2010

DOE – From Fundamental Science to Technology Research



DEPARTMENT OF ENERGY

Federal Energy Regulatory Commission

Office of the Secretary
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Daniel B. Poneman, Deputy Secretary*

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- Biological & Environmental Research
- Fusion Energy Science
- High Energy Physics
- Nuclear Physics
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- Assistant Secretary for Policy & International Affairs
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- Intelligence and Counterintelligence

- Energy Information Administration
- Southeastern Power Administration
- Bonneville Power Administration
- Western Area Power Administration
- Southwestern Power Administration

* The Deputy Secretary also serves as the Chief Operating Officer

26 May 09

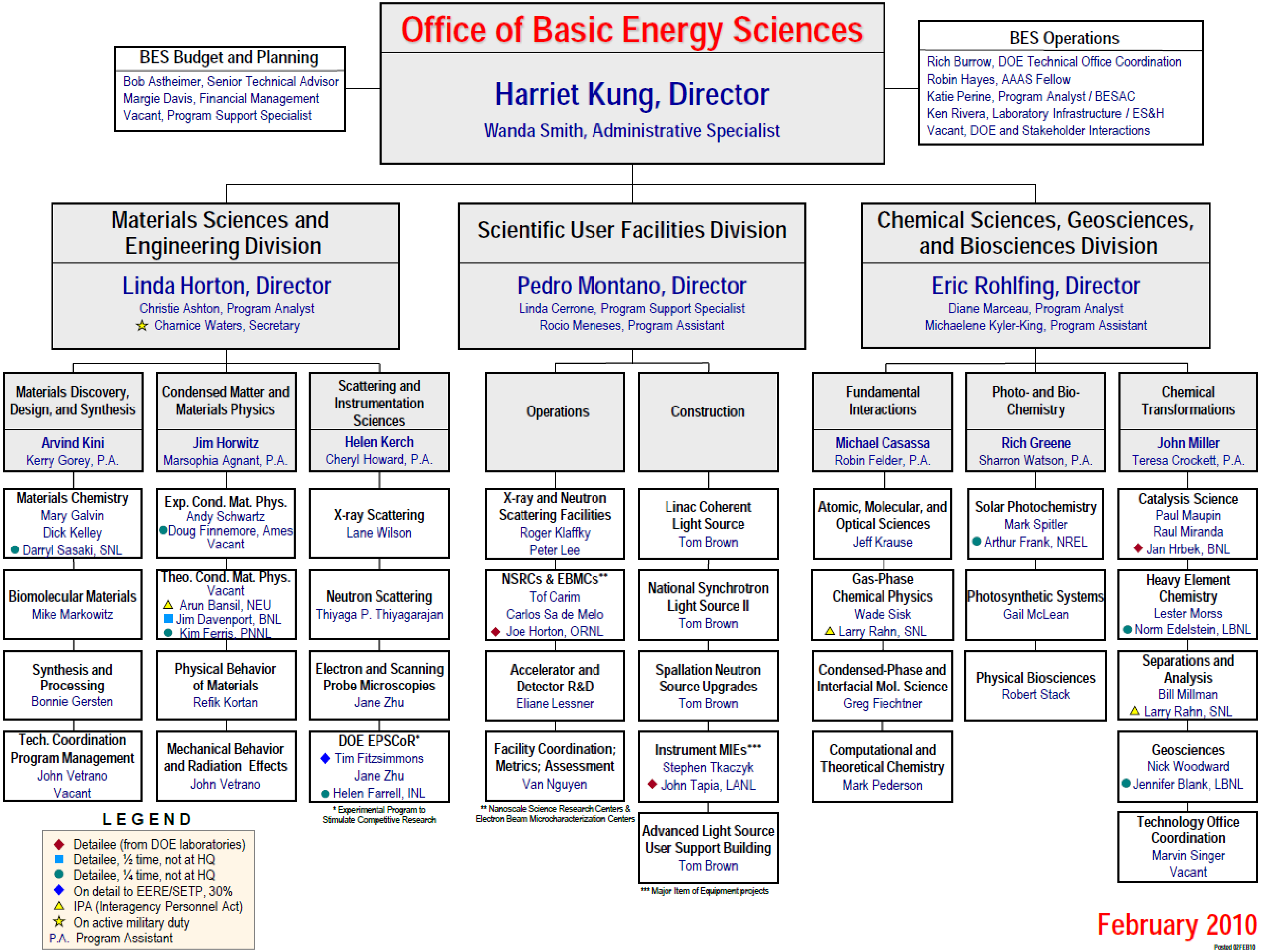
Basic Energy Sciences Mission

Mission:

- Fundamental research to understand, predict, and ultimately control matter and energy at the electronic, atomic, and molecular levels
- Provide the foundations for new energy technologies to support DOE's missions in energy, environment, and national security
- Plan, construct, and operate world-leading scientific user facilities for the Nation

Priorities:

- Discover and design new materials and molecular assemblies with novel function, through atom-by-atom and molecule-by-molecule control
- Conceptualize, calculate, and predict processes underlying physical and chemical transformations
- Probe, understand, and control the interactions of phonons, photons, electrons, and ions with matter to direct and control energy flow in materials and chemical systems
- To foster integration of the basic research with research in the DOE technology programs and NNSA



Office of Basic Energy Sciences

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Wanda Smith, Administrative Specialist

BES Budget and Planning
Bob Astheimer, Senior Technical Advisor
Margie Davis, Financial Management
Vacant, Program Support Specialist

BES Operations
Rich Burrow, DOE Technical Office Coordination
Robin Hayes, AAAS Fellow
Katie Perine, Program Analyst / BESAC
Ken Rivera, Laboratory Infrastructure / ES&H
Vacant, DOE and Stakeholder Interactions

Materials Sciences and Engineering Division

Linda Horton, Director

Christie Ashton, Program Analyst
☆ Charnice Waters, Secretary

Materials Discovery, Design, and Synthesis
Arvind Kini
Kerry Gorey, P.A.

- **Materials Chemistry**
  Mary Galvin
  Dick Kelley
  ● Darryl Sasaki, SNL
- **Biomolecular Materials**
  Mike Markowitz
- **Synthesis and Processing**
  Bonnie Gersten
- **Tech. Coordination Program Management**
  John Vetrano
  Vacant

Condensed Matter and Materials Physics
Jim Horwitz
Marsophia Agnant, P.A.

- **Exp. Cond. Mat. Phys.**
  Andy Schwartz
  ●Doug Finnemore, Ames
  Vacant
- **Theo. Cond. Mat. Phys.**
  Vacant
  △ Arun Bansil, NEU
  ■ Jim Davenport, BNL
  ● Kim Ferris, PNNL
- **Physical Behavior of Materials**
  Refik Kortan
- **Mechanical Behavior and Radiation Effects**
  John Vetrano

Scattering and Instrumentation Sciences
Helen Kerch
Cheryl Howard, P.A.

- **X-ray Scattering**
  Lane Wilson
- **Neutron Scattering**
  Thiyaga P. Thiyagarajan
- **Electron and Scanning Probe Microscopies**
  Jane Zhu
- **DOE EPSCoR***
  ♦ Tim Fitzsimmons
  Jane Zhu
  ● Helen Farrell, INL

* Experimental Program to Stimulate Competitive Research

Scientific User Facilities Division

Pedro Montano, Director

Linda Cerrone, Program Support Specialist
Rocio Meneses, Program Assistant

Operations

- **X-ray and Neutron Scattering Facilities**
  Roger Klaffky
  Peter Lee
- **NSRCs & EBMCs****
  Tof Carim
  Carlos Sa de Melo
  ◆ Joe Horton, ORNL
- **Accelerator and Detector R&D**
  Eliane Lessner
- **Facility Coordination; Metrics; Assessment**
  Van Nguyen

** Nanoscale Science Research Centers & Electron Beam Microcharacterization Centers

Construction

- **Linac Coherent Light Source**
  Tom Brown
- **National Synchrotron Light Source II**
  Tom Brown
- **Spallation Neutron Source Upgrades**
  Tom Brown
- **Instrument MIEs*****
  Stephen Tkaczyk
  ◆ John Tapia, LANL
- **Advanced Light Source User Support Building**
  Tom Brown

*** Major Item of Equipment projects

Chemical Sciences, Geosciences, and Biosciences Division

Eric Rohlfing, Director

Diane Marceau, Program Analyst
Michaelene Kyler-King, Program Assistant

Fundamental Interactions
Michael Casassa
Robin Felder, P.A.

- **Atomic, Molecular, and Optical Sciences**
  Jeff Krause
- **Gas-Phase Chemical Physics**
  Wade Sisk
  △ Larry Rahn, SNL
- **Condensed-Phase and Interfacial Mol. Science**
  Greg Fiechtner
- **Computational and Theoretical Chemistry**
  Mark Pederson

Photo- and Bio-Chemistry
Rich Greene
Sharron Watson, P.A.

- **Solar Photochemistry**
  Mark Spitler
  ● Arthur Frank, NREL
- **Photosynthetic Systems**
  Gail McLean
- **Physical Biosciences**
  Robert Stack

Chemical Transformations
John Miller
Teresa Crockett, P.A.

- **Catalysis Science**
  Paul Maupin
  Raul Miranda
  ◆ Jan Hrbek, BNL
- **Heavy Element Chemistry**
  Lester Morss
  ● Norm Edelstein, LBNL
- **Separations and Analysis**
  Bill Millman
  △ Larry Rahn, SNL
- **Geosciences**
  Nick Woodward
  ● Jennifer Blank, LBNL
- **Technology Office Coordination**
  Marvin Singer
  Vacant

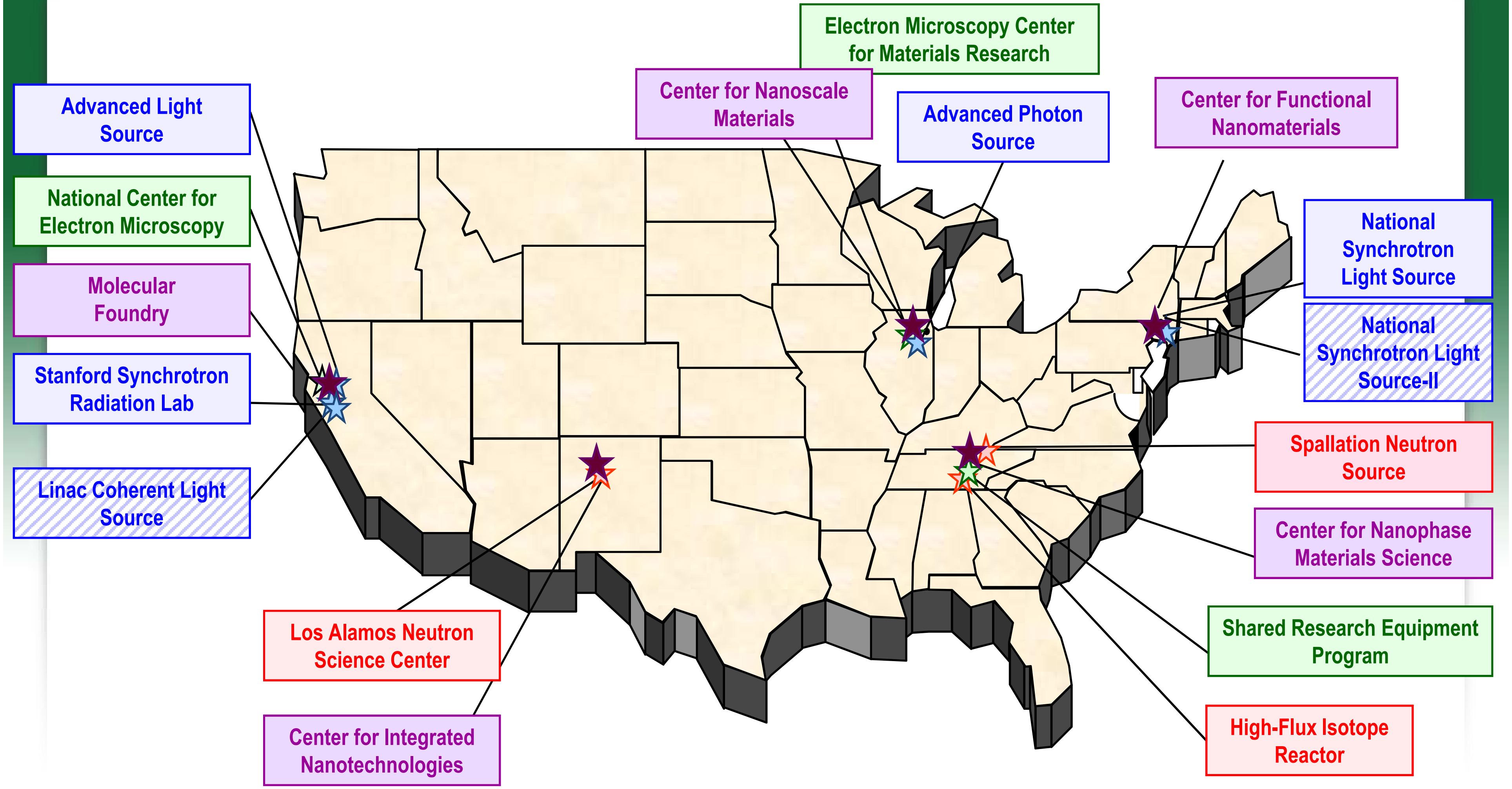
LEGEND

◆ Detailee (from DOE laboratories)
■ Detailee, ½ time, not at HQ
● Detailee, ¼ time, not at HQ
♦ On detail to EERE/SETP, 30%
△ IPA (Interagency Personnel Act)
☆ On active military duty
P.A. Program Assistant

February 2010

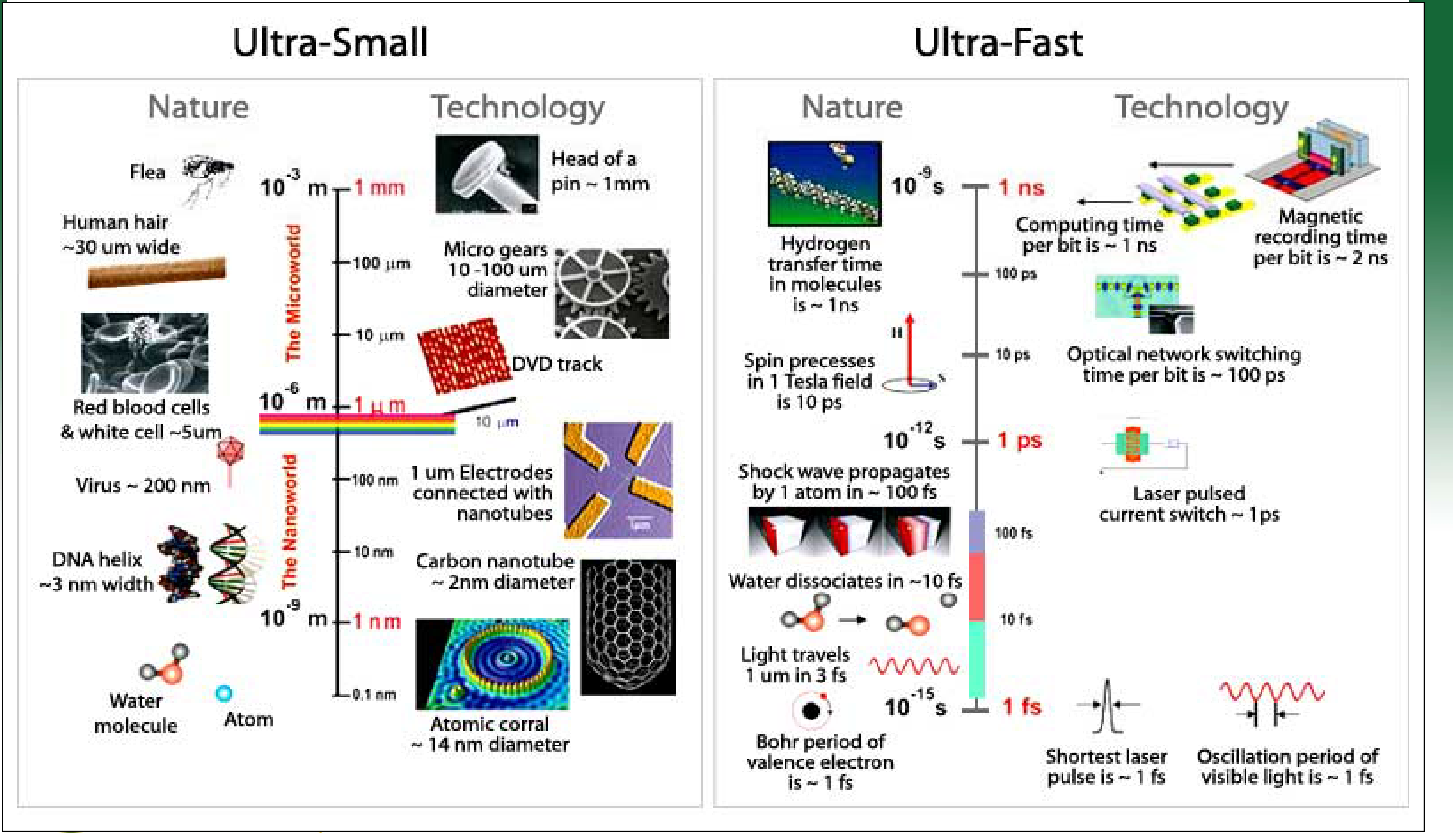
Posted 02FEB10

BES Scientific User Facilities: Resources for Materials Research



- 4 Synchrotron Radiation Light Sources
- Linac Coherent Light Source (Under construction)
- 3 Neutron Sources
- 3 Electron Beam Microcharacterization Centers
- 5 Nanoscale Science Research Centers

Ultra-small and Ultra-fast:
Frontiers in Science & Technology



Strategic Planning at BES

Opportunities Expressed in The "Basic Research Needs …" Workshop Series

Identifying Basic Research Directions for Today's and Tomorrow's Energy Technologies

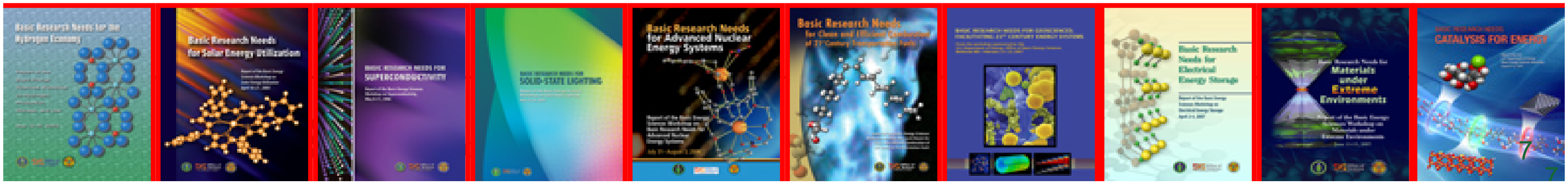
Basic Research Needs for a Secure Energy Future (BESAC) - 2003


BASIC RESEARCH NEEDS
TO ASSURE
A SECURE ENERGY FUTURE

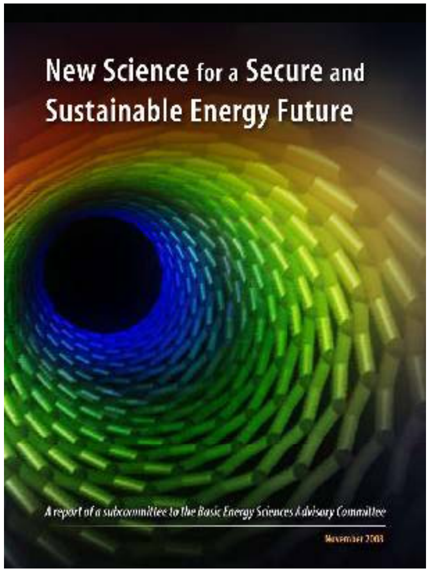
A Report from the
Basic Energy Sciences Advisory Committee

http://www.sc.doe.gov/bes/reports/files/SEF_rpt.pdf

- Basic Research Needs for the Hydrogen Economy
- Basic Research Needs for Solar Energy Utilization
- Basic Research Needs for Superconductivity
- Basic Research Needs for Solid State Lighting
- Basic Research Needs for Advanced Nuclear Energy Systems
- Basic Research Needs for the Clean and Efficient Combustion of 21st Century Transportation Fuels
- Basic Research Needs for Geosciences: Facilitating 21st Century Energy Systems
- Basic Research Needs for Electrical Energy Storage
- Basic Research Needs for Catalysis for Energy Applications
- Basic Research Needs for Materials under Extreme Environments



Additional Workshops and Resources



New Science for a Secure and Sustainable Energy Future

A report of a subcommittee to the Basic Energy Sciences Advisory Committee

November 2003

New Science for a Secure and Sustainable Energy Future

Goals:

- Make fuels from sunlight
- Generate electricity without carbon dioxide emissions
- Revolutionize energy efficiency and use

Recommendations:

- Work at the intersection of control science and complex functional materials
- Increase the rate of discoveries
- Establish “dream teams” of talent, equipped with forefront tools, and focused on the most pressing challenges to increase the rate of discovery
- Recruit the best talent through workforce development to inspire today’s students and young researchers to be the discoverers, inventors, and innovators of tomorrow’s energy solutions

How Nature Works ··· to ··· Materials and Processes by Design to ··· Technologies for the 21st Century

Grand Challenges

How nature works

- Controlling materials processes at the level of quantum behavior of electrons
- Atom- and energy-efficient syntheses of new forms of matter with tailored properties
- Emergent properties from complex correlations of atomic and electronic constituents
- Man-made nanoscale objects with capabilities rivaling those of living things
- Controlling matter very far away from equilibrium

Discovery and Use-Inspired Basic Research

Materials properties and functionalities by design

- Basic research for fundamental new understanding on materials or systems that may revolutionize or transform today's energy technologies
- Development of new tools, techniques, and facilities, including those for the scattering sciences and for advanced modeling and computation
- Basic research, often with the goal of addressing showstoppers on real-world applications in the energy technologies

Applied Research

- Research with the goal of meeting *technical milestones*, with emphasis on the development, performance, cost reduction, and durability of materials and components or on efficient processes
- Proof of technology concepts

Technology Maturation & Deployment

- Scale-up research
- At-scale demonstration
- Cost reduction
- Prototyping
- Manufacturing R&D
- Deployment support

BESAC & BES Basic Research Needs Workshops

BESAC Grand Challenges Report

DOE Technology Office/Industry Roadmaps

Basic Energy Sciences
Goal: new knowledge / understanding
Mandate: open-ended
Focus: phenomena
Metric: knowledge generation

DOE Technology Offices: EERE, NE, FE, EM, RW…
Goal: practical targets
Mandate: restricted to target
Focus: performance
Metric: milestone achievement

Budget for Hydrogen-related Research at BES

Hydrogen research is not a line-item request in the BES budget but funding for hydrogen-related research has been tracked internally in a consistent fashion since 2005.

BES Funding for hydrogen research

•FY2008	$36.4 M
•FY2009	$38.7 M
•FY2010	$38.7 M

Increases in FY2009 are a result of new hydrogen-related Energy Frontier Research Centers and several proposals funded under the "Single Investigator and Small Group Research" (SISGR) program

Emphasis

Continued focus on critical basic research needs for hydrogen production, storage, and use:

- Hydrogen Storage
- Membranes
- Nanoscale Catalysts
- Solar Hydrogen Production
- Bio-Inspired Hydrogen Production

Basic Sciences Underpinning Technology

- **Coordination between basic science and applied research and technology is an important mechanism by which to translate transformational discoveries into practical devices**

- **Many activities facilitate cooperation and coordination between BES and the technology programs**
  - Joint efforts in strategic planning (e.g., 10 BRN workshops)
  - Solicitation development
  - Reciprocal staff participation in proposal review activities
  - Joint program contractors meetings
  - Joint SBIR topics
  - Participation by BES researchers at the Annual Merit Review

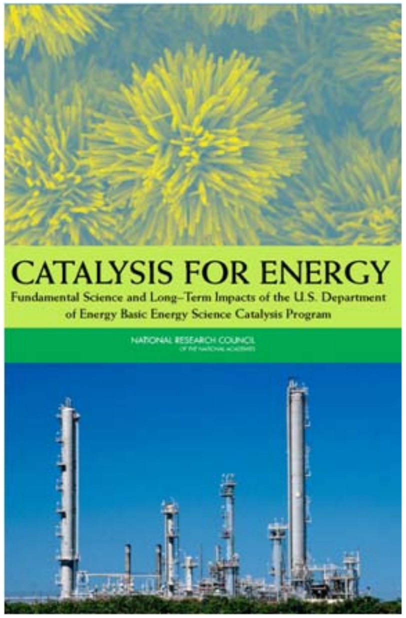
- **Co-funding and co-siting of research by BES and DOE technology programs at DOE labs or universities, has proven to be a viable approach to facilitate close integration of basic and applied research through sharing of resources, expertise, and knowledge of research breakthroughs and program needs.**

Platinum Monolayer Electrocatalysts: BES-EERE-Industry

Brookhaven National Laboratory

1. Use-inspired BES research on electrochemical interfaces leads to discovery of a new class of nano-catalysts.
2. The EERE fuel cell program supports the development of the new catalysts for fuel cell applications.
3. Industrial support via CRADAs demonstrate synthesis scale up and excellent performance in fuel cell tests.
4. New BES research now turns to catalysts for ethanol fuel cells.

- BES user facilities – the NSLS and Center for Nanofunctional Materials (CFN) – provided key characterization capabilities (x-ray absorption spectroscopy and advanced microscopy).
- This work was featured as one of the 10 most impactful research efforts in the NAS review of the BES Catalysis Science Program.



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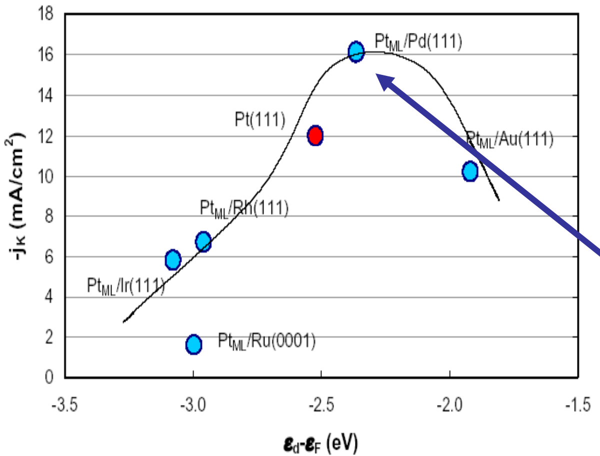
Radoslav Adzic and co-workers, Chemistry Department,
Brookhaven National Laboratory

Platinum Monolayer Electrocatalysts

BES-supported research 1992-present at Brookhaven National Laboratory

Fundamental studies of electrocatalysis:
Oxygen reduction reaction (ORR) – mechanism, structure/activity

Insight (2000): Platinum monolayers are promising catalysts

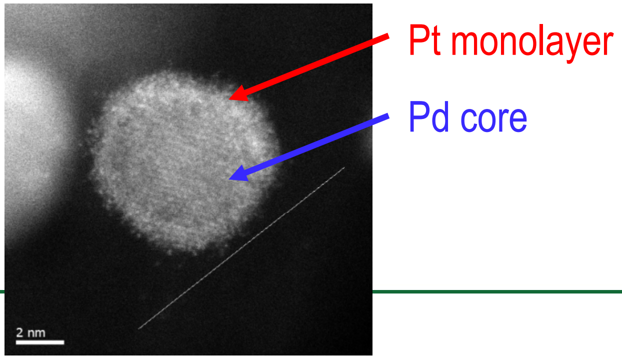


Substrate tunes catalytic activity of Pt overlayer:
- -Catalytic activity correlates to O binding energy
- -Optimum at intermediate binding: volcano plot
- -Stability can also be tuned

Pt monolayer on Pd(111) high ORR activity

Strategy: address critical cost and stability limits of fuel cell ORR catalysts

Nanostructured core-shell electrocatalysts
- - active monolayer puts all Pt atoms at interface
- - substrate core tunes activity & stability



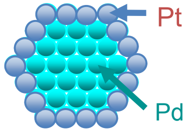
Platinum Monolayer Electrocatalysts

EERE-supported research 2003-present at Brookhaven National Laboratory

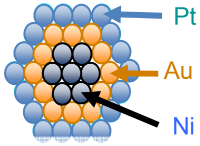
1. Several classes: tune properties & reduce cost
2. Atomic-level characterization *in situ* (XAS), *ex situ* (STEM), DFT
3. Atomic-level control syntheses fine-tune Pt-core interactions and control morphology
4. Activity, Stability and Fuel Cell tests

<ins>Catalytic Activity</ins> improved 5x-20x per wt Pt

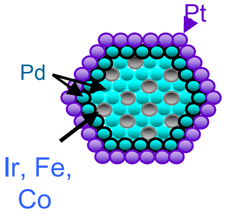
<ins>Durability improved:</ins> multiple thousand hours in LANL tests



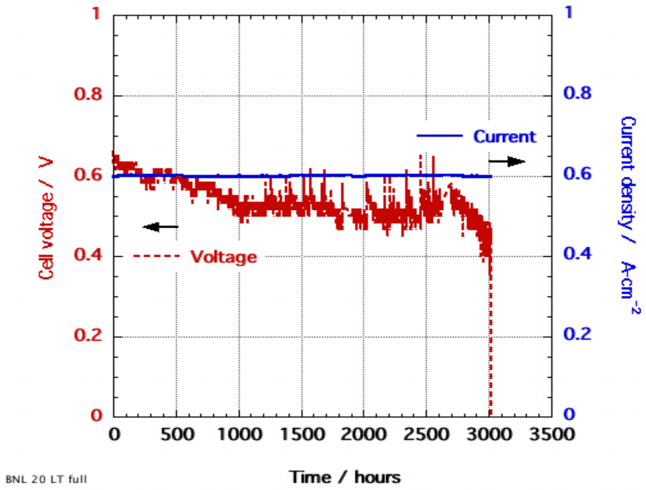
Pt monolayer on Pd nanoparticles



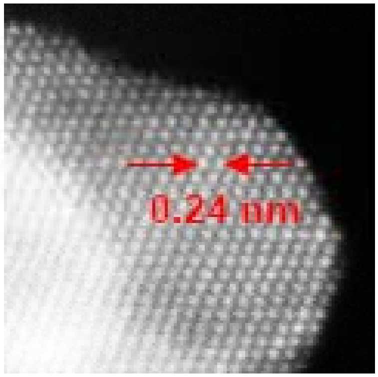
Pt on non-noble metal – noble metal core-shell;

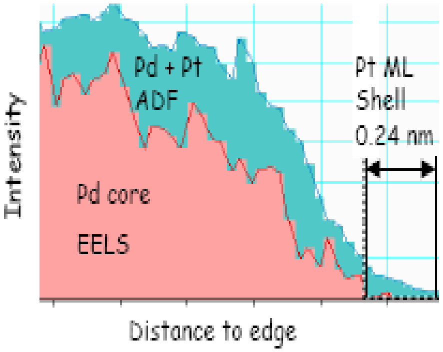


Pt on nanoparticles with interlayer of Pd



<ins>Characterization:</ins> atomic imaging of one monolayer of Pt shell on Pd-core nanoparticles using STEM/EELS at Center for Functional Nanomaterials





Platinum Monolayer Electrocatalysts

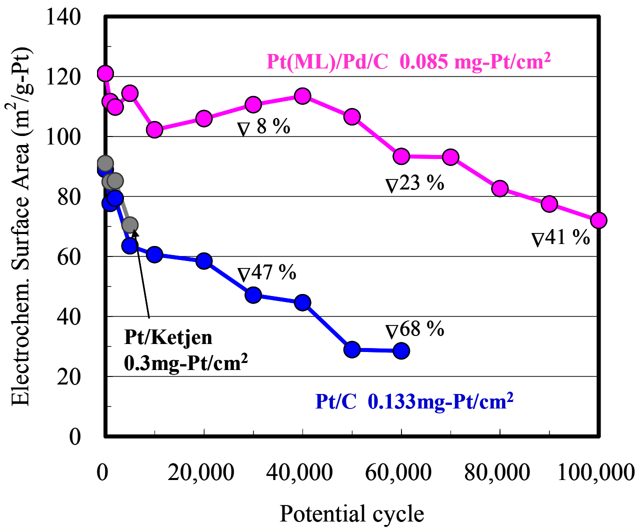
CRADAs with industry 2005-present at Brookhaven National Laboratory

Scale-up, fuel cell testing: Toyota, GM, UTC Fuel Cells, Battelle

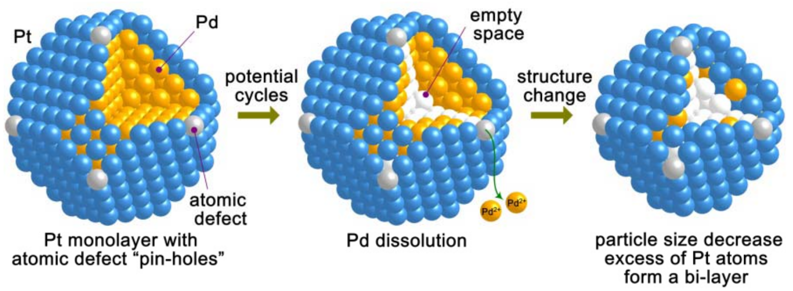
1. Demonstrate efficient, reproducible synthesis of gram quantities of Pt_{ML}
2. Fuel cell tests, performance, stability, potential cycling

<u>Synthesis:</u> e.g., Demonstrate scale-up to 50 gram batches of high-activity nanostructured core-shell electrocatalysts (with Toyota).

<u>Performance:</u> High activity, improved stability in MEA-level cycling. Scale-up will enable full fuel cell stack testing.



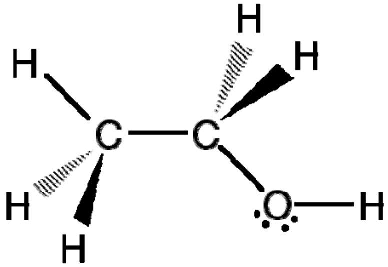
<u>Understand improved stability:</u> Evidence that Pd core acts as ‘sacrificial electrode’ for Pt shell.



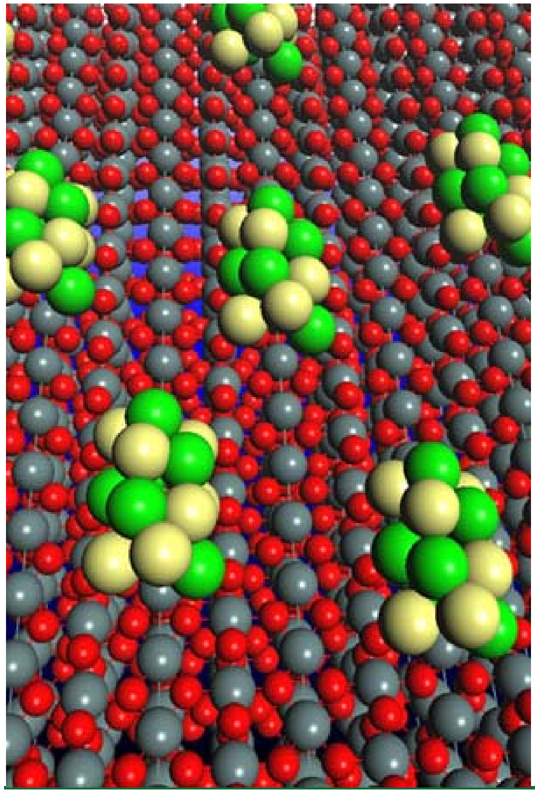
<u>Status:</u> Core-shell nanocatalysts currently promising route to PEM fuel cell commercialization.

New BES research on catalysts for ethanol fuel cells

Brookhaven National Laboratory



Challenge: *stable, selective and energy-efficient C-C oxidation in a fuel cell with fuel molecules containing C-C, C-O, C-H bonds.*



- Model of a ternary electrocatalyst for ethanol oxidation consisting of platinum-rhodium clusters on a surface of tin dioxide. For the first time, this catalyst can split the carbon-carbon bond selectively at a fuel cell anode.

- Hydrogen adsorbate binds through the hollow Rh-Pt site, all other species bind through the bare Rh sites; the cluster structure forces the formation of a cyclic intermediate that results in C-C bond breakage.

BES-supported critical advance: use of characterization techniques (EXAFS, IRRAS) at NSLS and CFN and molecular modeling techniques (DFT) to understand the role of bimetallic cluster structure, support structure, electronic structure, and charge transfer on the mechanism of C-C splitting and oxidation.

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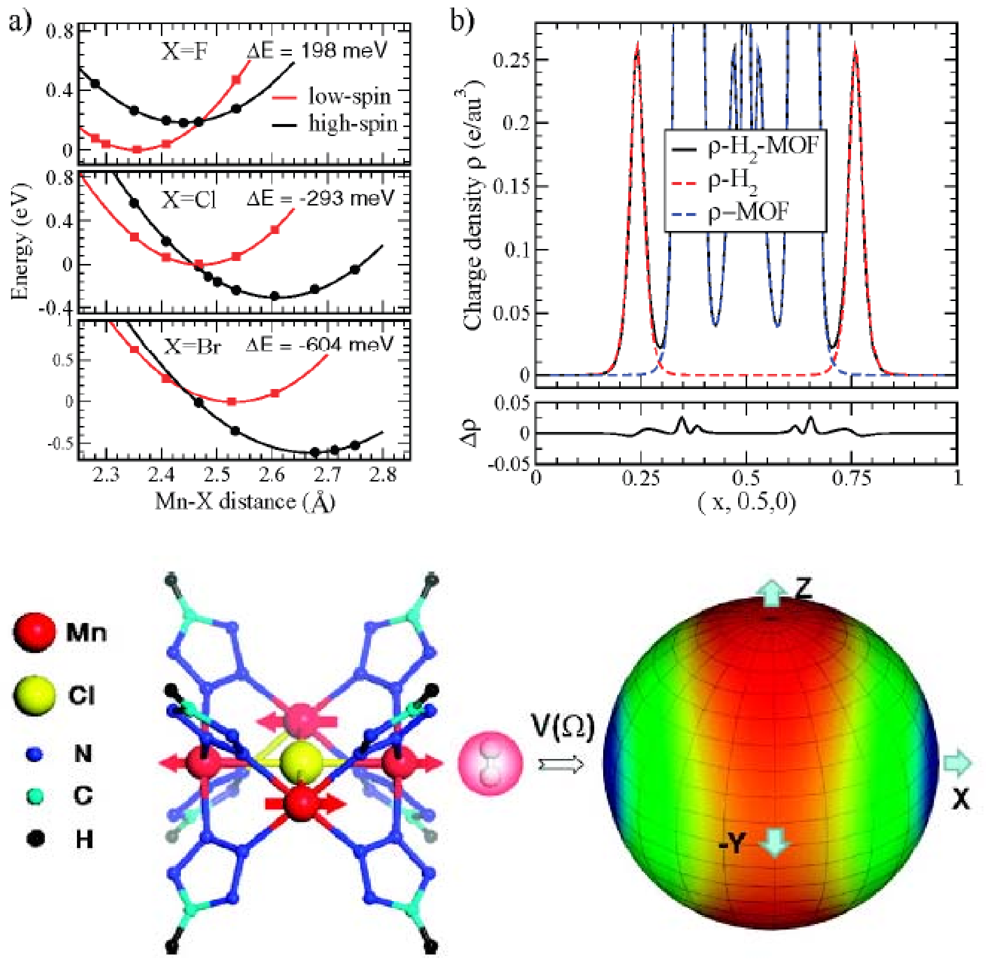
R. Adzic, et al., Nature Materials 8, 325 (2009)

Tunability of Hydrogen Binding in Metal-Organic Frameworks with Exposed Transition Metal Sites

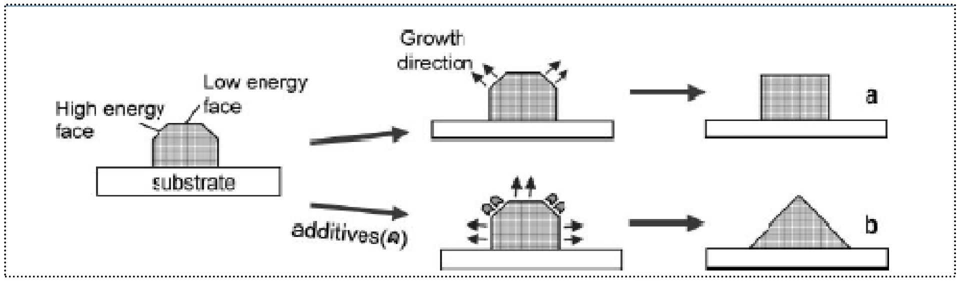


W. Zhou and T. Yildirim, *J. Phys. Chem. C* 112 (22) 2008

- Recent results incorporating exposed transition metals (TM) sites in Mn_4Cl-MOFs have shown an increase in binding energy (BE) to a level intermediate between pure van der Waals (4 kJ/mol) and Kubas binding (30-50 kJ/mol) but the mechanism was not clear and originally attributed to a type of Kubas interaction
- BE was shown to depend on the magnetic spin state of the Mn ions and that this spin state could be influenced by exchanging Cl with either F or Br, either reducing or increasing the BE, respectively
- DFT calculations were made on these systems that showed the hydrogen-TM binding was not a Kubas-type but was rather a Coulomb interaction (no electron sharing nor bond stretching) with little hybridization of orbitals
- This Coulomb interaction is very anisotropic and to properly calculate the binding energy it is necessary to take into account the quantum nature of the H2 orientation. Here it is shown that the rotational dynamics of the H2 molecule are strongly confined to a slab-like region (red region in bottom right figure) which has been shown to influence the BE.

Novel Synthesis of Nanomaterials for Solar Hydrogen Production

- Cu_2O is a direct-gap semiconductor and is a viable candidate to produce hydrogen by direct water photolysis using visible light with little or no external bias. The efficiency of photocurrent generation is dependent on the shape, size and interconnection of the polycrystals.
- By gaining a deeper understanding of the growth mechanisms of electrodeposited Cu_2O crystals it is possible to control the growth in such a way as to tailor the final shape and size of the crystals and therefore alter the generation of photocurrent when the crystals are illuminated.



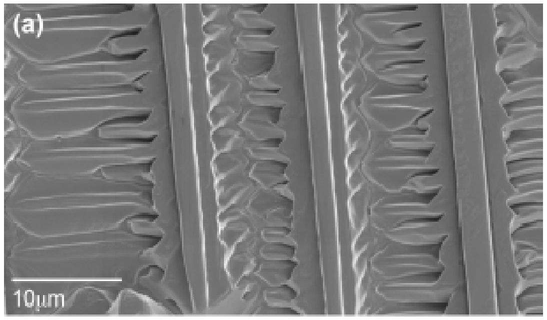
Cu_2O →

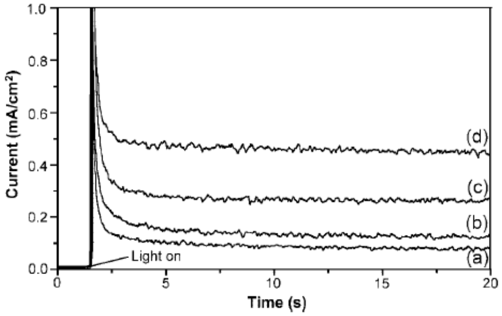

Dalton Transactions

An international journal of inorganic chemistry

RSCPublishing

- Two keys to branching morphology were found to be the buffering of the solution to eliminate localized changes in pH and the control of overpotential during growth; precise manipulation of these two factors allows exquisite control of dendritic branching.
- Once this was fully understood it was possible, by creating optimally branched structures, to increase the photocurrent of a thin film of Cu_2O by up to a factor of 20, thereby increasing the ability to produce hydrogen from sunlight.

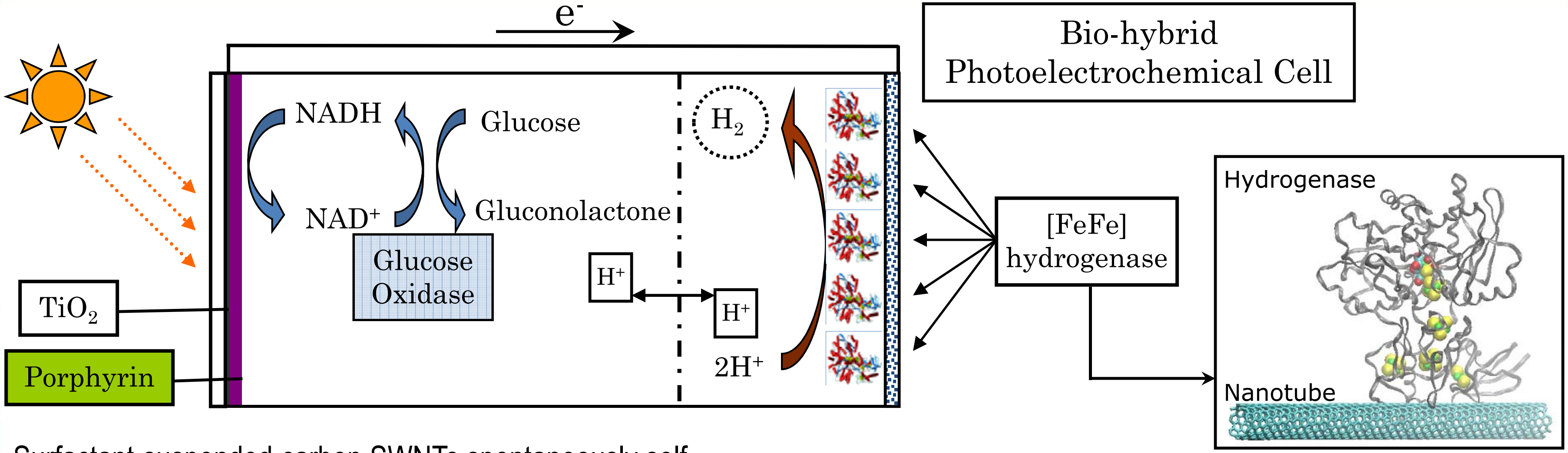




K.-S. Choi, *Dalton Trans.*, 2008, p5432

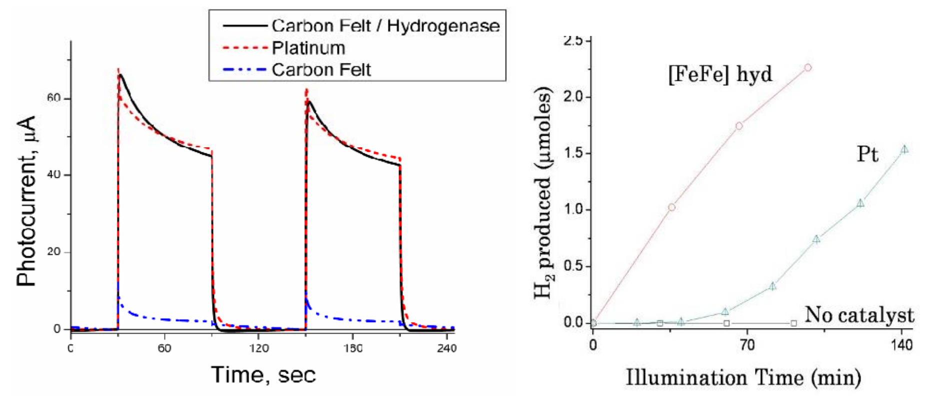
C.M. McShane and K.-S Choi, *J. Am. Chem. Soc.*, 2009, 131 (7) p. 2561

Bio-hybrid H_2-production with [FeFe] hydrogenase



- Surfactant-suspended carbon SWNTs spontaneously self-assemble with [FeFe] hydrogenases to form catalytically active biohybrids
- SWNTs act as molecular wires, making electrical contact to the biocatalytic region of the hydrogenase
- [FeFe] hydrogenase immobilized onto carbon electrodes in a PEC serves as a model Bio-hybrid, Solar-driven, H_2-production system
- H_2-production photocurrents, rates and durations with [FeFe] hydrogenase as catalyst closely match the performance values of a nanoparticulate, Pt-catalyst

Comparable performance of [FeFe] hydrogenase and



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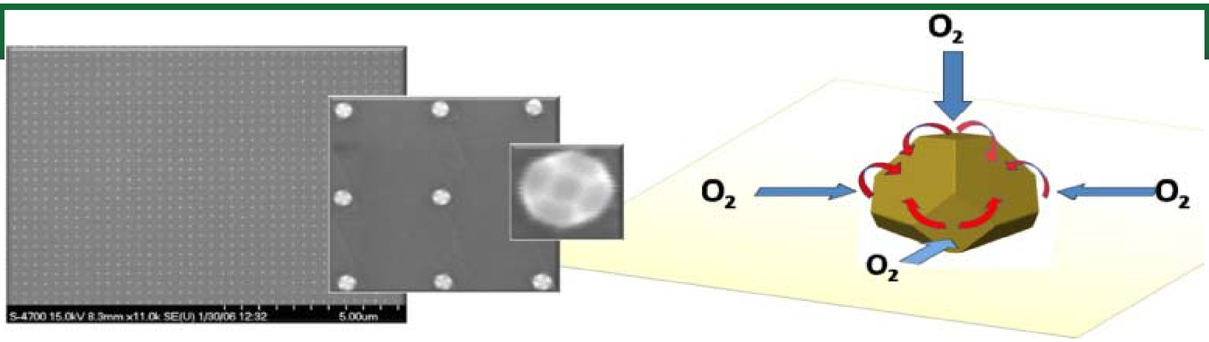
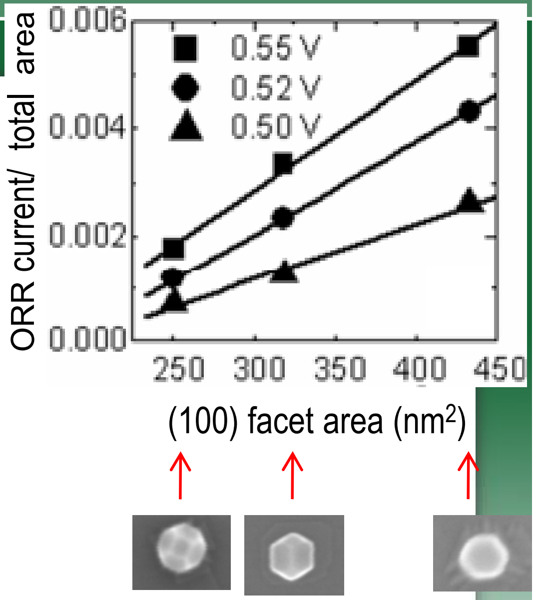
T.J. McDonald et al, NanoLetters 7 (11) 2007

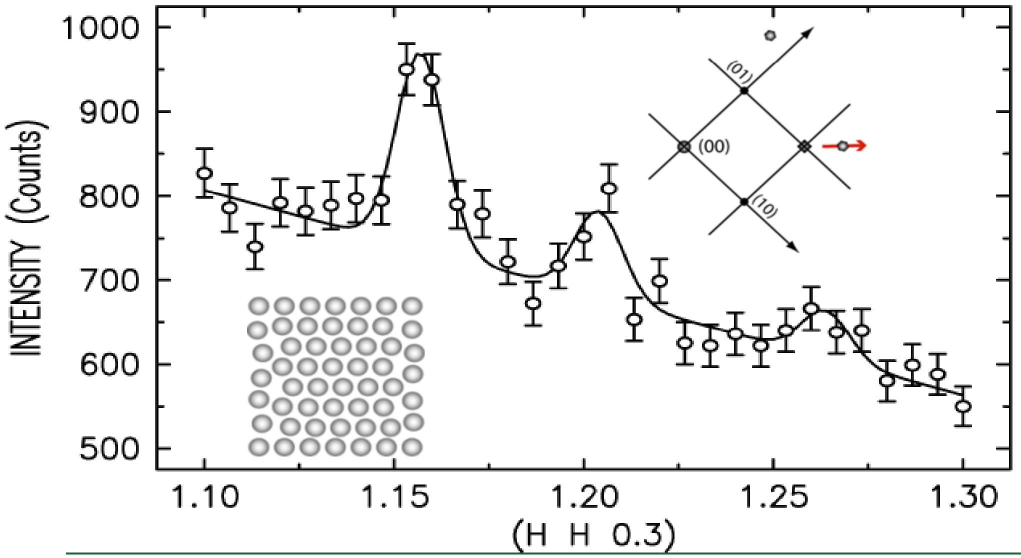
M.C. Beard, J.L. Blackburn and M.J. Heben, NanoLetters 8 (12) 2008

Catalytic Mechanisms Elucidated with Pt Nanoparticle Arrays



Arrays of replicated Pt nanoparticles were tested in $HClO_4$ for oxygen reduction reaction (ORR) activity. Distinct substrate orientations produced particles with differing ratios of (111) to (100) facet area. Each array exhibits nanoparticles of nearly identical size, shape, and orientation.



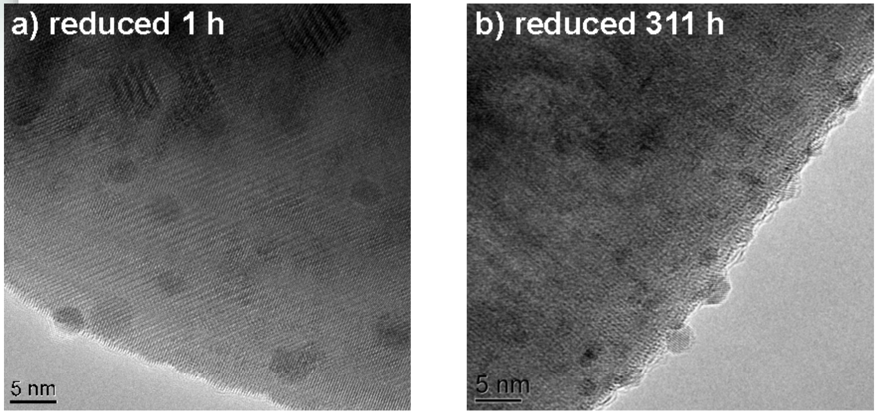


- The (100) nanofacet is found by synchrotron x-ray experiments to have partial ‘hex’-surface reconstruction, known for inducing catalytic activity in large (100) surfaces.
- ORR is proportional to the area of 1st step adsorbing (100) facets, while activation energies match those of the final steps found on active (111) surfaces.
- High activity of the particles are explained by nanoscale crossover of intermediates.

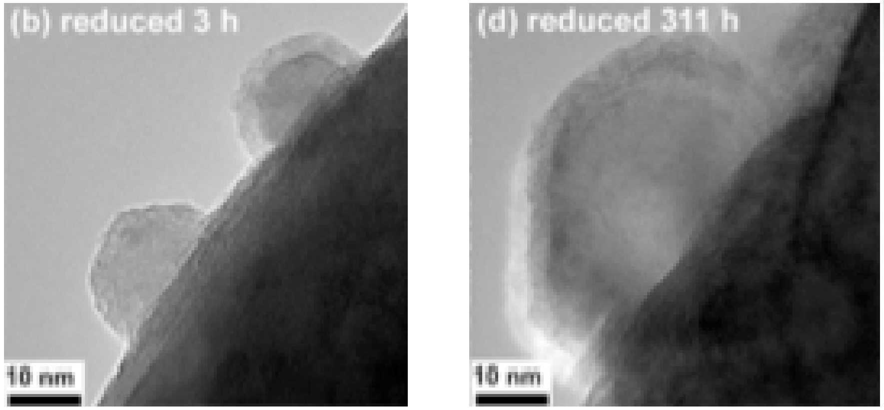
V Komanicky, H Iddir, K-C Chang, A Menzel, G Karapetrov, D Hennessy, P Zapol, H You, *J. Am. Chem. Soc.* **131**, 5732 (2009)

Dynamics of Nano-Catalyst Precipitation in (La,Sr)(Cr,M)O_3 Composite Anodes

- New anode materials are important for making fuel-flexible solid oxide fuel cells
- Novel anodes have been developed where nano-sized catalyst particles form on oxide surfaces
  - Example, Ru particles form on (La,Sr)(Cr,Ru)O_3 surfaces, improving electrical performance, when they are exposed to the reducing fuel during cell operation
  - The particles show remarkable stability even at temperatures of 800°C (lower left image)
- To help understand these results, a new system, (La,Sr)(Cr,Ni)O_3, was studied
- Ni forms much larger particles than Ru, that grow substantially over time at 800°C (lower right)
  - Differences explained by the lower melting point of Ni, which may lead to a higher surface diffusivity on the oxide particles



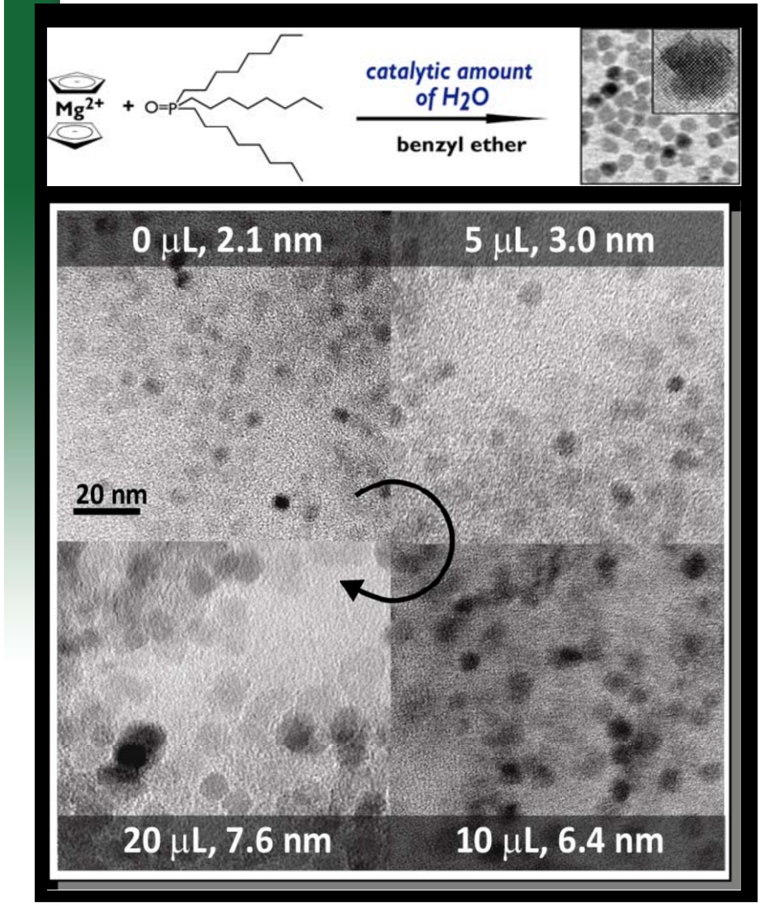
Electron microscope images of Ru particles on oxide surfaces, that are very small (2-5 nm) and remain stable in the reducing fuel environment



Electron microscope images of Ni particles on oxide surfaces, that are much larger than Ru and grow rapidly in the reducing fuel environment

Kobsiriphat; Madsen; Wang; Shah; Marks; Barnett; J. Electrochem Soc 2010, 157, B279

Size-Controlled Monodisperse Colloidal Magnesium Oxide Nanocrystals: Synthesis and Efficient Blue Luminescence

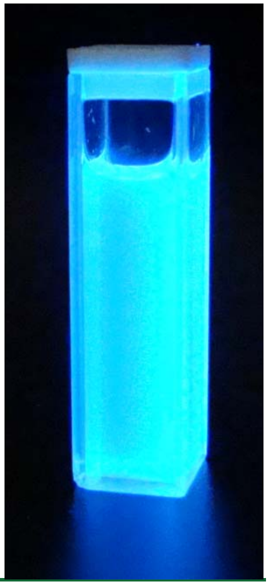


- Monodisperse nanocrystals of MgO can be prepared by direct colloidal synthesis. Diameter control (2-8 nm) is achieved by incorporating catalytic amounts of water into the organic reaction medium.

- Bright, blue luminescence is observed with quantum yield up to ca. 20%. The large population of surface states on these small nanocrystals is responsible for the efficient blue emission, which is unexpected from a wide band-gap insulator.

- MgO nanocrystals could play a role in long-term storage of carbon dioxide, a potential means of crucial tactic in tempering the effects of global warming.

- *MgO provides a introductory route to development of Mg particles for true hydrogen storage.*



Angew. Chem. Int. Ed. (2009), *48*, 6278, *Selected by the editor as a "hot" paper*
Highlighted by Reuters and Photonics Spectra (Long and Urban, LBNL)

Energy Frontier Research Centers

Tackling Our Energy Challenges in a New Era of Science

- To engage the talents of the nation's researchers for the broad energy sciences
- To accelerate the scientific breakthroughs needed to create advanced energy technologies for the 21st century
- To pursue the fundamental understanding necessary to meet the global need for abundant, clean, and economical energy

46 centers awarded ($777M over 5 years), representing 102 participating institutions in 36 states and D.C.

Pursue *collaborative* basic research that addresses both energy challenges and science grand challenges in areas such as:

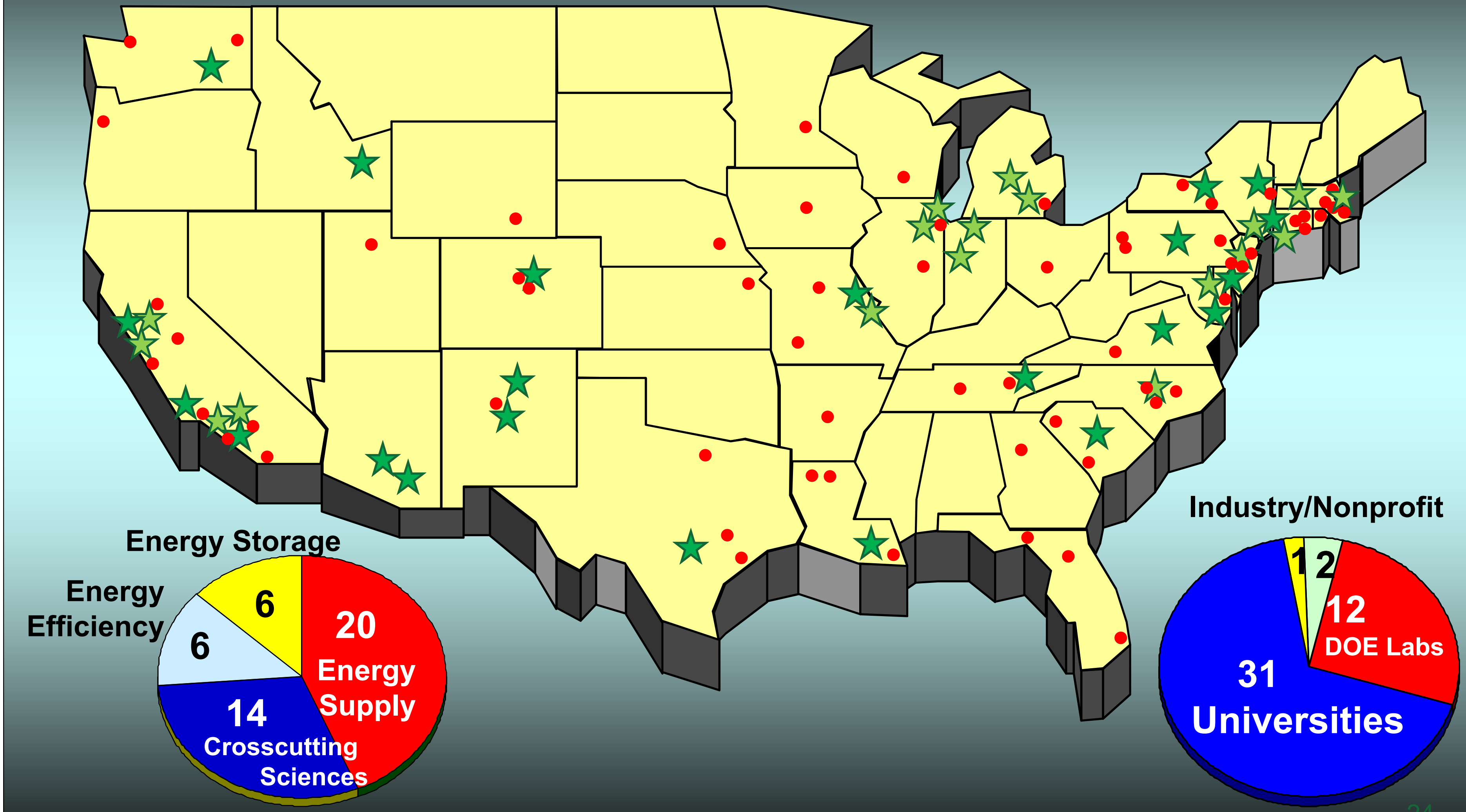
- Solar Energy Utilization
- Combustion
- Bio-Fuels
- Catalysis
- Energy Storage
- Solid State Lighting
- Geosciences for Energy Applications
- Superconductivity
- Advanced Nuclear Energy Systems
- Materials Under Extreme Environments
- Hydrogen

Summaries of all EFRCs available at http://www.sc.doe.gov/bes/EFRC/index.html

Energy Frontier Research Centers

46 centers awarded, representing 102 participating institutions in 36 states plus D.C

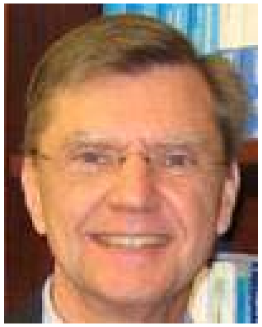
Energy Frontier Research Center Locations (★ Leads; ● Participants)



By Topical Category

By Lead Institution

Examples of Hydrogen-Related EFRCs



Michael Wasielewski, Northwestern Univ.

Argonne-Northwestern Solar Energy Research Center (ANSER

Tom Meyer, Univ. of North Carolina

Solar Fuels and Next Generation Photovoltaics

Ken Reifsnider, Univ. of South Carolina

Heterogeneous Functional Materials Center (HeteroFoaM)

Héctor Abruña, Cornell Univ.

Energy Materials Center at Cornell (EMC^2)

Energy Innovation Hub – Fuels from Sunlight

Part of a significant new DOE initiative - one of three Hubs funded at $22M each in the FY 2010 appropriation.

Key Dates:

- December 22, 2009: Funding Opportunity Announcement (FOA) issued
- January 29, 2010: Letters of intent due (but not required)
- March 29, 2010: Full applications due
- June, 2010: Award announcement
- August 2010: Award initiation

The objective of the Fuels from Sunlight Hub is to develop an effective solar energy to chemical fuel conversion system. The system should operate at an overall efficiency and produce fuel of sufficient energy content to enable transition from bench-top discovery to proof-of-concept prototyping.

For information on DOE Energy Innovation Hubs see: http://www.hubs.energy.gov/