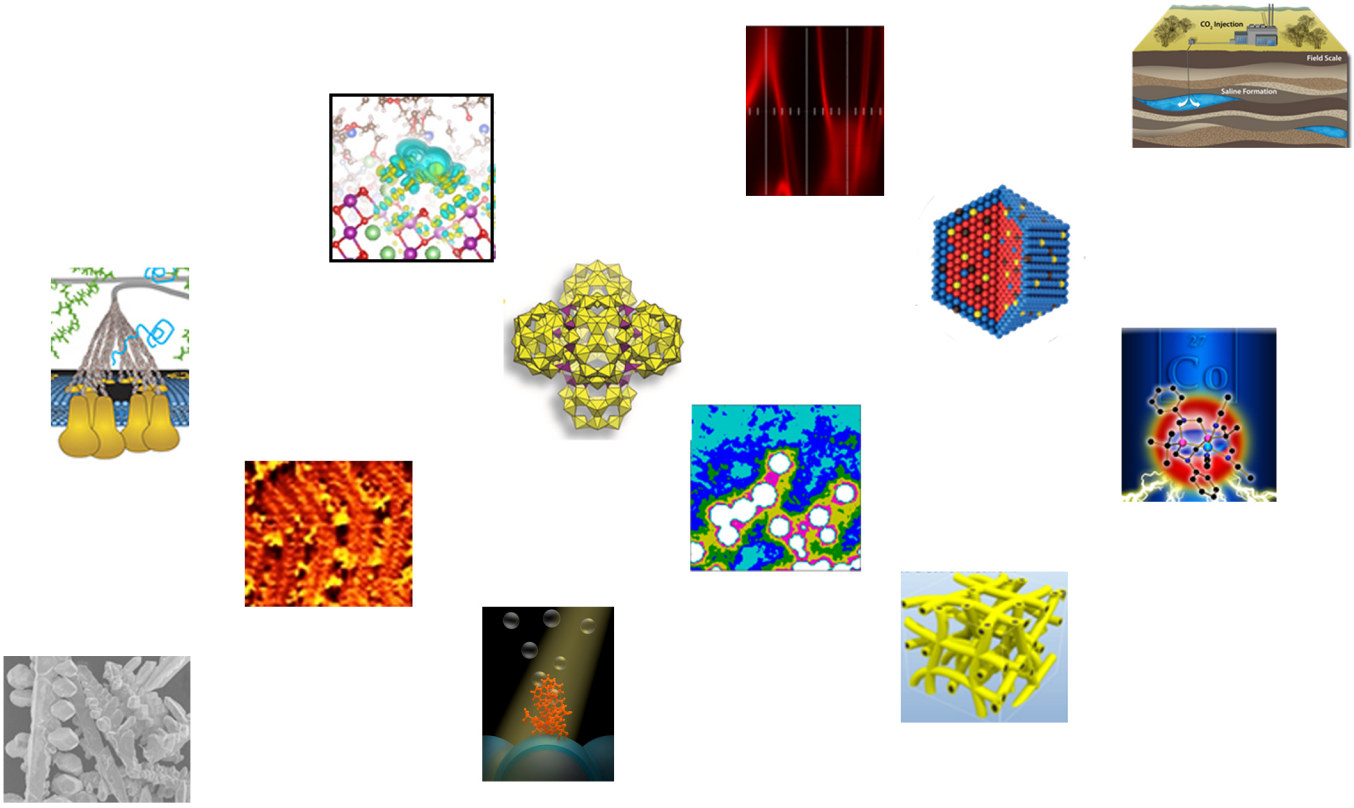
Energy Frontier Research Centers

Technical Summaries



August 2014
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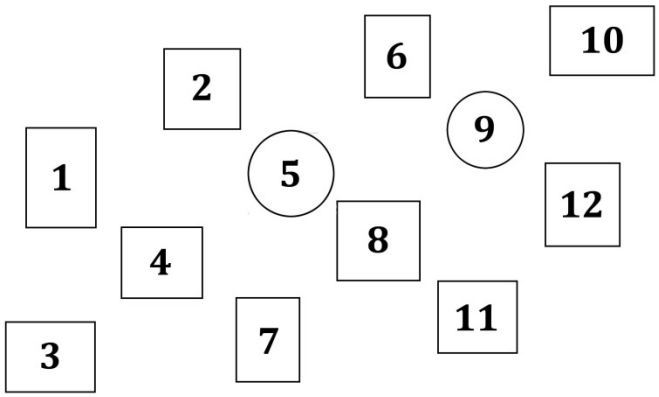

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INTRODUCTION

Since its inception in 2009, the U. S. Department of Energy's Energy Frontier Research Center (EFRC) program has become an important research modality in the Department's portfolio, enabling high impact research that addresses key scientific challenges for energy technologies. Funded by the Office of Science's Basic Energy Sciences program, the EFRCs are located across the United States and are led by universities, national laboratories, and private research institutions. These multi-investigator, multi-disciplinary centers bring together world-class teams of researchers, often from multiple institutions, to tackle the toughest scientific challenges preventing advances in energy technologies. The EFRCs' fundamental scientific advances are having a significant impact that is being translated to industry.

In 2009 five-year awards were made to 46 EFRCs, including 16 that were fully funded by the American Recovery and Reinvestment Act (ARRA). An open recompetition of the program in 2014 resulted in four-year awards to 32 centers, 22 of which are renewals of existing EFRCs and 10 of which are new EFRCs. The technical research summaries in this document describe the planned work of these 32 EFRCs and list all the partner institutions and associated senior investigators. The index at the end of the document includes keywords, grand challenges, and references to BES reports that the leadership of each EFRC has identified as relevant to their centers.

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Center for Electrochemical Energy Science (CEES)
EFRC Director: Paul Fenter
Lead Institution: Argonne National Laboratory
Start Date: August 2009

Mission Statement*: To create a robust fundamental understanding of the phenomena that control the reactivity of electrified oxide interfaces, films and materials relevant to lithium-ion battery chemistries.*

Energy storage is a strategic technology that enables technologies ranging from the large-scale use and distribution of electrical energy to smaller-scale applications for mobile technologies, for example, vehicles and portable electronic devices. These applications place strong demands on the performance of electrical energy storage systems in terms of their ability to provide energy and power in a safe manner. Energy storage technologies rely on the reversible conversion of electrical and chemical energy through electrochemical reactions. These reactions include ion transport between the cathode and anode (and associated electron transfer reactions), as well as deleterious side reactions that can occur between the electrolyte and the electrodes. Lithium-ion batteries, in particular, are attractive because they offer inherently high energy densities. Nevertheless, much remains unknown about the molecular structures and the dynamic processes at the electrode-electrolyte interface that control the electrochemically driven reactions.

The *Center for Electrochemical Energy Science* (CEES) seeks to create a robust, fundamental understanding of electrochemically-driven reactivity using lithium-ion battery chemistry as a well-defined model system. Our primary goal is to understand the reactivity of electrified oxide materials, films and interfaces that can be leveraged to control nano- and mesoscale interactions. Oxide materials represent many of the positive electrode materials found in energy storage systems. These systems have a richness and a complexity that result from the large number of variables controlling their electrochemical properties (e.g., vacancies, cation and anion substitution and ordering, and redox potentials that can be manipulated by composition, crystal orientation, lattice strain and temperature). A robust molecular-scale understanding of these complex processes will enable the discovery of fundamentally new concepts, structures and architectures that, ultimately, may lead to disruptive advances in energy storage systems that involve electrified materials and interfaces.

To this end, CEES seeks to answer two broad questions that will guide the Center's research (indicated schematically in **Fig. 1**).

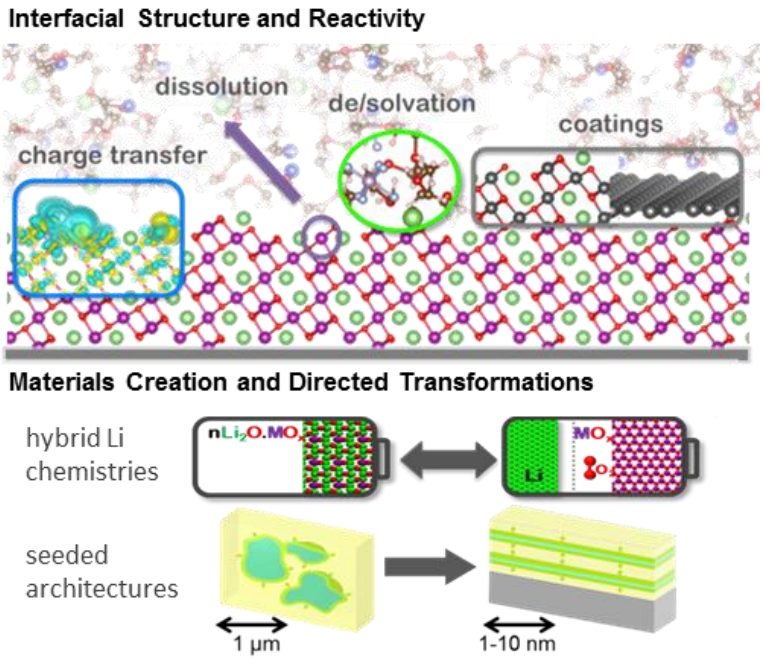

Figure 1: Interfacial and materials structures and processes that will be studied in CEES, including the observation and control of interfacial reactivity and the direction of chemical reactivity through advanced materials and novel materials.

- **Interfacial Structure and Reactivity**: *Can we control, and autonomically respond to, reactions at the oxide-electrolyte interface by leveraging a robust molecular-scale understanding of its structure and reactivity?* The transport of ions across the electrode/electrolyte interface leads to kinetic barriers, over-potentials, and side reactions associated with interface-specific reaction pathways, for example, in the "solid electrolyte interphase" (SEI) layer. This is exemplified by the well-known problem of oxide instabilities at extreme potentials. These interfacial aspects of electrochemistry introduce substantial challenges, but simultaneously offer many avenues for controlling and guiding reactivity through interfacial modification.
- **Materials Creation and Directed Transformations:** *Can we create new materials and discover new approaches that can be used to direct the character and nature of electrochemical reactions within electrode materials?* While the electrochemical reactions are thermodynamically controlled, the pathway of an electrochemically-driven reaction within an electrode can be influenced. For example, 1) the dimensionality of nano-structured electrodes can be used to influence the initiation of lithiation reactions in a material, and 2) dual functioning electrode/electrocatalysts materials can enable reversible lithium and oxygen extraction from a lithium-transition metal-oxide crystal structure while allowing partial redox of the transition metal ions.

These guiding questions will be addressed using the proven multi-pronged approach that CEES has established and used over the past 5 years. To this end, CEES will probe the intrinsic reactivity of well-defined model oxide systems, with molecular-scale sensitivity and resolution, to reveal the inherent complexity of electrochemical systems (including the relevant active materials, electrolyte, etc.). The use of precisely defined materials, structures and interfaces (for example, structurally and compositionally defined thin films) will enable direct observations of the relevant reactions without the complications of inactive materials and complex electrode morphologies inherent to conventional systems. This understanding, leveraged with novel materials, architectures and chemistries, will be used to observe and direct electrochemical reactivity in these systems. These approaches, enabled by our deep cross-cutting capabilities of materials synthesis, characterization, and theory will lead to new approaches to control electrochemical reactions under the extreme conditions typically found in lithium-ion battery systems.

Center for Electrochemical Energy Science (CEES)	
Argonne National Laboratory	Paul Fenter (Director), Michael Thackeray (Deputy Director), Khalil Amine, Maria Chan, Larry Curtiss (Task 1 Lead), Jeffrey Elam (Task 2 Lead), Timothy Fister, Christopher Johnson
Northwestern University	Mark Hersam (Institutional Lead) , Scott Barnett, Michael Bedzyk, Vinayak Dravid, Tobin Marks, Christopher Wolverton
University of Illinois at Urbana-Champaign	Andrew Gewirth (Institutional Lead) , Ralph Nuzzo, Nancy Sottos, Scott White
Purdue University	Jeffrey Greeley

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Northeast Center for Chemical Energy Storage (NECCES)
EFRC Director: M. Stanley Whittingham
Lead Institution: Binghamton University
Start Date: August 2009

Mission Statement*: To develop an understanding of how key electrode reactions occur, and how they can be controlled to improve electrochemical performance, from the atomistic level to the macroscopic level through the life-time of the operating battery.*

The design of the next generation of rechargeable batteries requires both the development of new chemistries and the fundamental understanding of the physical and chemical processes that occur in these complex systems. Although some significant advances have been made to prepare and utilize new materials, efforts towards the understanding of mechanisms have waned. This will eventually choke efforts to efficiently develop new materials if this issue is not addressed now. Batteries are inherently complex and dynamic systems, their electrochemistry, phase transformations, and transport processes often varying throughout their lifetime. Although often viewed as simple to use by the customer, their successful operation relies heavily on a series of complex mechanisms, involving thermodynamic instability in many parts of the charge- discharge cycle and the formation of metastable phases. The requirements for long-term stability are extremely stringent and necessitate control of the chemistry at a wide variety of temporal and structural length scales. This in turn necessitates the development and use of new characterization tools to monitor these processes. The overall goal is to understand the transformations (and their rates) that occur in an electrode composite structure, from the atomistic level to the macroscopic level, throughout the lifetime of the functioning battery. The four-year scientific research goals are:

1. Close the gap between the theoretical and practical energy density for intercalation compounds.
2. Attain reversible multi-electron transfer in a cathode material using lithium.
3. Understand performance limiting transport in positive electrode structures from the local through the meso to the macroscale.
4. *Enable new chemistries* involving electrode systems that were previously considered intractable for use in batteries.

These research goals will be achieved by dividing our research effort into three closely connected and integrated thrusts; the theory effort is integrated into thrusts 1 and 2.

Thrust 1: Intercalation Materials Chemistry. This thrust will identify the key parameters that are required to optimize intercalation reactions in the active material in the electrodes. We will determine:

(i) The structural parameters required for rapid ionic (Li^+) motion (and thus high rate), particularly at close to full lithium removal
(ii) The minimal electronic (and ionic conductivity), required to completely extract Li from materials for different crystal structures and particle sizes.
(iii) How to control (minimize) structural changes so as to enable redox processes involving more than one electron
(iv) The key structural parameters required to enable battery chemistries involving ions other than Li^+.

Thrust 2. Transport - Establishing the Local-Meso-Macro Scale Continuum. This thrust will establish a comprehensive understanding of the ionic and electronic transport in model electrode materials and establish a direct link to electrochemical performance through the correlation of physical phenomena in the increasingly complex hierarchy of a model battery electrode. We will determine:

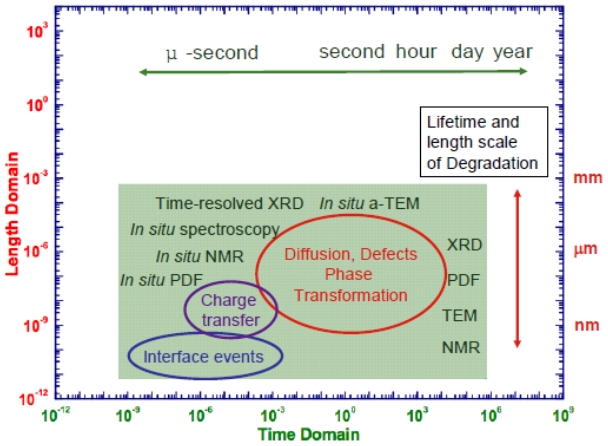
(i) Definitive links of local ionic and electronic transport to correlated physical phenomena occurring across and within phase transformations.

(ii) The complex ionic and electronic pathways at the meso to microscale to develop a working theory of composite electrode design.

Thrust 3: Cross-cutting research: Developing the characterization and diagnostic tools to investigate battery function. This thrust will involve the development of novel *in-* and *ex- situ* experimental approaches aimed at probing electrical energy storage (EES) materials at three levels: atom, single crystal/particle, and across the electrode heterostructure. An emphasis is placed on *in-situ* methods that use multiple experimental tools simultaneously or that combine imaging with spectroscopy. *In-situ* spectroscopy is crucial as the materials are exceptionally dynamic over all levels of material structure during the operation of a battery and can be highly sensitive to ambient contamination if *ex-situ* techniques are employed. Metastable phases, including key reaction intermediates can relax to form different phases if probed by ex-situ techniques. For the three levels discussed above, several major developments are proposed:

(i) *Atomic level structure:* operando XAS, diffraction, PDF, SAXS, and NMR, annular dark/bright field STEM imaging of both heavy and light elements.

(ii) *Single crystal/particle level imaging:* Strain mapping with CXDI, high energy resolution STEM-EELS; and Nano-scale mapping at the nm-level (at the Debye length) across interfaces – including oxidation state mapping, in situ TEM, high energy resolution STEM-EELS.



(iii) *Imaging of reactions across the electrodes heterostructure:* Imaging the whole electrode intact ideally under operando conditions, through development of high resolution PDF/SAXS tomography and compatible operando electrochemical cells; structural insights with NMR experiments of studies of transport connected with MRI, PFG and SIMS studies of structure and tortuosity.

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Rutgers University	G. Amatucci (Assoc. Director, Thrust 2 leader), F. Cosandey
University of Cambridge	C.P. Grey
Massachusetts Institute of Technology	G. Ceder, Y-M. Chiang
Argonne National Laboratory	K. Chapman (Thrust 3 leader), P. Chupas
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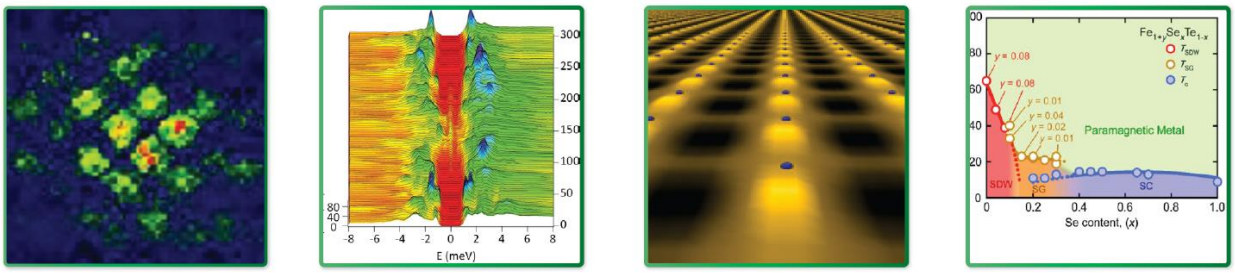
Center for Emergent Superconductivity (CES)
EFRC Director: Peter D. Johnson
Lead Institution: Brookhaven National Laboratory
Start Date: August 2009

Mission Statement*: To discover new high-temperature superconductors and improve the performance of known superconductors by understanding the fundamental physics of superconductivity.*

As U.S. electrical energy consumption continues to grow, the nation's electrical power transmission grid faces fundamental structural challenges of capacity, reliability and efficiency if it is to meet the needs of the 21st century. Electricity demand will grow by 50% in the US and by 100% globally by 2030, with nearly all of that growth in cities and suburbs where the overhead power lines and underground cables are already saturated. Power delivery and control solutions based on superconductors could solve these crises by using their demonstrated higher current carrying capacities over conventional cables, self-healing fault current limiting capabilities, and substantial increases in efficiency. However, there remain many fundamental materials and physics challenges which must be addressed in order for superconductivity to have broad impact on the electrical grid.



From left to right: Experimental evidence for Charge Density Wave structure in cuprate superconductor discovered inside a vortex core using scanning tunneling microscopy (STM); Spectra taken at many points across the vortex core; Artist's rendition of the cuprate nematic phase; Phase diagram for the iron based superconductor $FeTe_{1-x}Se_x$ as a function of the doping level x.

The objective of the *Center for Emergent Superconductivity* (CES) is to explore fundamental research issues with the objective to overcome key barriers leading to the viable application of high temperature/high current superconductivity. This will be achieved by enabling the design of superconducting materials with optimal physical and critical properties for deployment of a 21th century superconducting power grid. Thus, the most profound challenge of CES is to understand the fundamental mechanisms of high-temperature and high-current superconductivity sufficiently so as to direct discovery of new or improved families of materials with higher critical temperatures and currents.

The central mission for the CES is the development of a broadly defined *Materials Genome Initiative* that encompasses three highly coordinated fundamental research directions specifically designed to work symbiotically towards the greater goal of the fundamental understanding high-temperature superconductivity. These are: a) the development of techniques to create of new classes of superconducting materials by design; b) a fundamental understanding of the mechanism of high-temperature superconductivity in existing materials, including the cuprates and Fe-based superconductors; and c) the fundamental understanding the current carrying limiting properties of

existing high-temperature superconductors that will lead to applications performance enhancement by design. We expect the unification of these priority research directions will prove transformative in our fundamental understanding and provide the basis for predictive design of new families of high-temperature superconductors.

Center for Emergent Superconductivity (CES)	
Brookhaven National Laboratory	Peter D. Johnson (Director) J. C. Séamus Davis, John M. Tranquada, Cedomir Petrovic, Ivan Bozovic, Genda Gu, Qiang Li, Mark Dean, Jon Rameau, John Hill
Argonne National Laboratory	Mike Norman, Wai Kwok, Alexei Koshelev, Duck-Young Chung, Dean Miller, Ulrich Welp, Mercouri Kanatzidis
University of Illinois, Urbana-Champaign	Laura Greene, David Ceperley, Peter Abbamonte, Jim Eckstein, Philip W. Phillips, Jian-Min Zuo
Rutgers University	Gabi Kotliar

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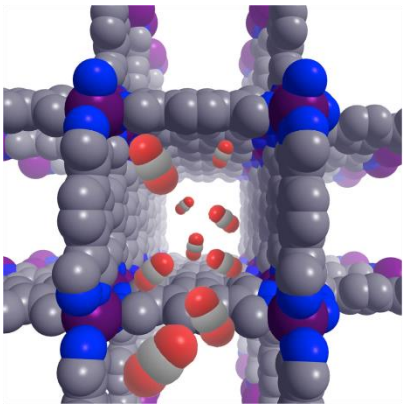
Center for Gas Separations Relevant to Clean Energy Technologies (CGS)
EFRC Director: Jeffrey R. Long
Lead Institution: University of California, Berkeley
Start Date: August 2009

Mission Statement*: To develop the fundamental science necessary for tailor-making materials and membranes for the efficient separation of gas mixtures, particularly as required in the clean use of fossil fuels and in the generation of alternative fuels.*

Separation processes are estimated to be responsible for 10-15% of our total energy consumption. Given the expected increase in global population and the possibility of large-scale carbon capture and sequestration, this percentage is anticipated to increase significantly. Reducing the total energy costs of separations would be a major contribution towards lowering our overall energy useage. The aim of the Center for Gas Separations (CGS) is to develop new strategies and materials that enable energy-efficient gas separations, based on molecule-specific chemical interactions, together with a fundamental understanding of the properties and performance of the materials.



The challenge is to remove the fundamental scientific barriers that currently prohibit the efficient separation of gases essential to the development of clean energy technologies. In gas separations these challenges are significant. The differences between the relevant gas molecules are small and therefore we need to use the type of molecular control that is offered by nanoscience and synthetic chemistry to tailor materials that have exactly the right adsorption and diffusion selectivity to enable an economic separation process.

The center brings together personnel with expertise in the following areas of emphasis:

- *Materials Synthesis*: The synthesis of new gas-permeable materials with control over the molecular functionalities to preferentially adsorb gas molecules is essential. Our focus here will be on: (i) generating novel metal-organic frameworks exhibiting molecule-specific chemical interactions and (ii) new membrane constructs incorporating these materials.
- *Materials Characterization*: Detailed atomic-level structural characterization of the new materials will be necessary both before and after exposure to gas samples in order to probe hypotheses on interaction mechanisms. In addition, accurate means of assessing the selectivity, kinetics, and thermodynamics of gas adsorbate binding will be needed to demonstrate efficacy and test computational models.
- *Computational Separations*: A strong computational component to the research will be essential for understanding the chemical interactions at a molecular level, as well as for guiding the synthetic efforts toward materials exhibiting high specificity and tunable interaction energies.

We aim to develop fundamental new means of synthesizing materials with tailored molecular interactions, while generating new options for energy-related gas separations, including the separation of CO_2 from power plant flue streams, the separation of CO_2 from natural gas deposits, the separation of oxygen from air, and the separation of various industrially relevant hydrocarbon mixtures.

Center for Gas Separations Relevant to Clean Energy Technologies (CGS)	
University of California, Berkeley	Jeffrey Long, Jeffrey Neaton, Jeffrey Reimer, Berend Smit, Ting Xu, Omar Yaghi
Lawrence Berkeley National Laboratory	Brett Helms, Jeffrey Kortright, David Prendergast, Wendy Queen, Simon Teat, Steve Whitelam
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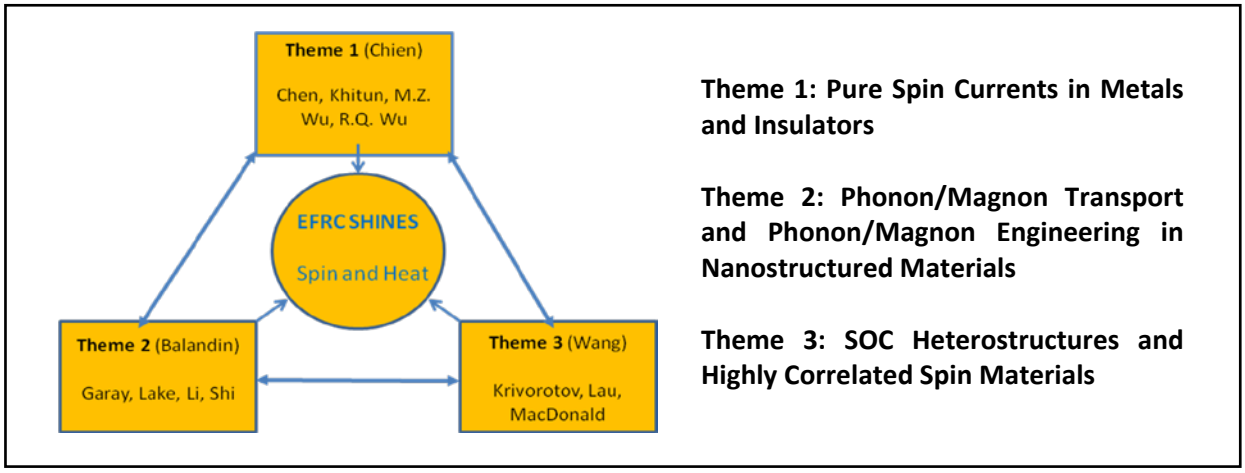
Spin and Heat in Nanoscale Electronic Systems (SHINES)
EFRC Director: Jing Shi
Lead Institution: University of California, Riverside
Start Date: August 2014

***Mission Statement**: To explore the interplay of spin, charge, and heat and to control the transport of spin and energy for achieving significantly higher energy efficiencies in nanoscale electronic devices.*

The four-year objectives for the SHINES center include (1) better understanding of and significant improvement in pure spin current effects in nanoscale electronic devices, including magnonic switching, spin-torque oscillations, spin-orbit torques, and spin Seebeck effect through novel materials and heterostructures; (2) engineering of acoustic phonon and magnon transport in nano-structured materials via controlling their dispersions and interactions; and (3) exploration of spin-orbit coupling for low energy effects and spin superconducting condensate for dissipationless spin and energy transport.

The SHINES center is organized in the following three interactive themes:

Theme 1: Pure spin currents in metals and insulators
This theme involves five members: Chien, Chen, Khitun, M.Z. Wu and R.Q. Wu and 5 cross-team members: Lake (Theme 2), Li (Theme 2), Shi (Theme 2), Wang (Theme 3), and Krivorotov (Theme 3). The theme is led by Chien. The main focus is on more efficient generation and detection of pure spin currents in metals and insulators and more efficient spin current-driven magnetization dynamics in both magnetic metal and insulator devices. The specific tasks include: high-precision determination of the spin Hall angle and spin diffusion length in strong spin-orbit coupling metals, especially ferromagnetic metals; search for large spin Hall angle and long spin diffusion length for high-efficiency energy



conversion from heat; improved understanding of the spin Hall angle and anomalous Hall effect in ferromagnetic metals and alloys; demonstration of spin torque oscillators (STO) in magnetic insulator devices based on the spin Seebeck effect; investigation of the Gilbert damping mechanism in doped and undoped magnetic insulator films; demonstration of magnonic switching in magnetic insulator-based thin film devices with point contact geometry; experimental investigation and micromagnetic simulations of magnonic switching dynamics; and measurements of propagating spin wave dynamics in magnetic insulator films and two-dimensional magnonic crystals.

Theme 2: Phonon/magnon transport and phonon/magnon engineering in nanostructured materials
This theme involves 5 members: Balandin, Garay, Lake, Li, and Shi, and 4 cross-theme members: M.Z. Wu (Theme 1), Khitun (Theme 1), Wang (Theme 3), and MacDonald (Theme 3). The theme is led by Balandin. The main focus of this theme is on the control of phonon/magnon properties to manipulate heat transport in nanostructured materials. The specific tasks include: demonstration of the acoustic phonon engineering effects on phonon dispersion and thermal transport (e.g. thermal conductivity) in nanostructured materials through material and structural property optimizations; demonstration of acoustic phonon spectrum modifications in spatially confined hertro- and nano-structures; fabrication and demonstration of two-dimensional magnonic crystals with band gaps via material and structural tuning; spectroscopic measurements of propagating chiral spin wave edge modes; determination of phonon/magnon temperature distributions in doped and undoped magnetic insulator films subjected to temperature gradient via Brillouin light scattering; investigation of physical mechanism of the spin Seebeck effect in magnetic insulators; demonstration of magnon/phonon Hall effect (i.e. anomalous Righi-Leduc effect) in magnetic insulator crystals; and experimental and theoretical investigations of the physical origin of the effect.

Theme 3: Spin-orbit coupling heterostructures and highly correlated spin materials
This theme involves 4 members: Wang, Krivorotov, Lau, and MacDonald, and 3 cross-theme members: Chien (Theme 1), Lake (Theme 2), and Balandin (Theme 2). The theme is led by Wang. The specific tasks include: demonstration and investigation of efficient spin-orbit torques in topological insulator/magnetic insulator heterostructures; theoretical and simulation studies of the effects of the spin-orbit torque on magnetization switching; demonstration and optimization of frequency-doubling high-efficiency spin torque oscillator nanodevices; better understanding of magnetization dynamics with micromagnetic simulations; measurements of spin-orbit coupling effects on electrical transport properties (e.g. weak localization and anti-localization) in transition metal dichalcogenide (TMD) nano-devices; measurements of temperature dependence of electron-phonon coupling in suspended TMD devices; theory and demonstration of spin superconductivity associated with the magnetic condensate in magnetic insulator nano-devices; and investigation of the stability and physical properties of the magnetic condensate.

Spin and Heat in Nanoscale Electronic Systems (SHINES)	
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University of California, Irvine	Ilya Krivorotov, Ruqian Wu
University of California, Los Angeles	Kang L. Wang
Johns Hopkins University	Chia-Ling Chien
Colorado State University	Mingzhong Wu
University of Texas at Austin	Elaine (Xiaoqin) Li, Allan MacDonald
Arizona State University	Tingyong Chen

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Light-Material Interactions in Energy Conversion (LMI)
EFRC Director: Harry Atwater
Lead Institution: California Institute of Technology
Start Date: August 2009

Mission Statement: *To tailor the morphology, complex dielectric structure, and electronic properties of matter so as to sculpt the flow of sunlight and heat, enabling light conversion to electrical and chemical energy with unprecedented efficiency.*

The Light-Material Interactions in Energy Conversion Energy Frontier Research Center (LMI-EFRC) is a national resource for fundamental optical principles and phenomena relevant to solar energy conversion, and for design of new photonic materials and structures used for energy conversion. This Center is a foundational partnership between scientific world leaders in optical properties of matter, internationally recognized experts in photovoltaic energy conversion, and innovators in the design and fabrication of novel electronic and photonic materials. The Center features a team of researchers that spans the campuses at Caltech, Harvard, Lawrence Berkeley National Laboratory, Stanford, and University of Illinois at Urbana-Champaign. In its renewal phase, the LMI-EFRC will comprehensively address new opportunities for very high efficiency solar energy conversion, making scientific discoveries that enable the efficient utilization of the entire visible and infrared solar resource.

Objectives for 2014-2018

Over the next four years, the overarching objective of the LMI-EFRC is to develop fundamental principles and new photonic materials and structures that can yield advances in ultrahigh solar conversion efficiency. Specifically, we aim to:

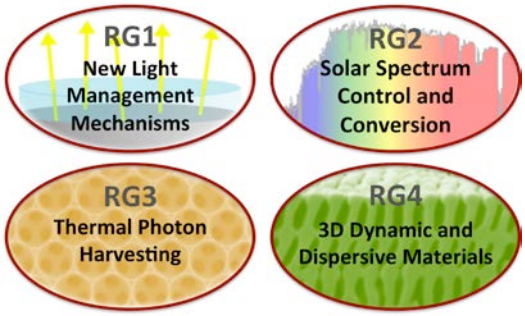
- Develop new mathematical methods for inverse design of light-management structures and mechanisms.
- Design photonic principles for structures enabling solar spectrum control to greatly enhance photovoltaic efficiency.
- Establish fundamental principles for thermal photon harvesting with a holistic view of both the emitter and absorber, and discover new structures and materials that enable thermal photon harvesting.
- Develop new light-directed and light-assisted methods of fabricating photonic materials and artificial photonic structures providing for passive and active control of light-matter interactions.

Selected Accomplishments to Date

- Developed flexible concentrator photovoltaics based on microscale crystalline silicon solar cells embedded in downshifting luminescent waveguides, increasing power output by greater than 300%.
- Discovered that to maximize photovoltaic conversion efficiency, photon emission from the solar cell should be maximized. Theoretical results led to Alta Devices' world record 27.6% efficient flat plate single-junction GaAs solar cell in 2011 and 28.8% in 2012 (and also 30.8% in 2014 for dual junction cell), exceeding the previous record by 2%.
- Developed light-driven material synthesis processes that enable energy conversion materials to develop their own complex architectures in response to illumination conditions.
- Established inverse design methods for optimization of light-trapping and dielectric spectrum-splitting photonic structures.
- Light-matter interaction principles and synthesis methods identified by LMI researchers led to technology developments by the companies Alta Devices, Caelux, and Semprius, MC10, and Electroninks Inc, all co-founded by LMI investigators.

Center Research Team and Scientific Organization

The Center is organized scientifically into four research groups (RGs) that address scientific themes related to light-matter interactions, with each team spanning multiple institutional partners and designed to address our four-year scientific objectives.



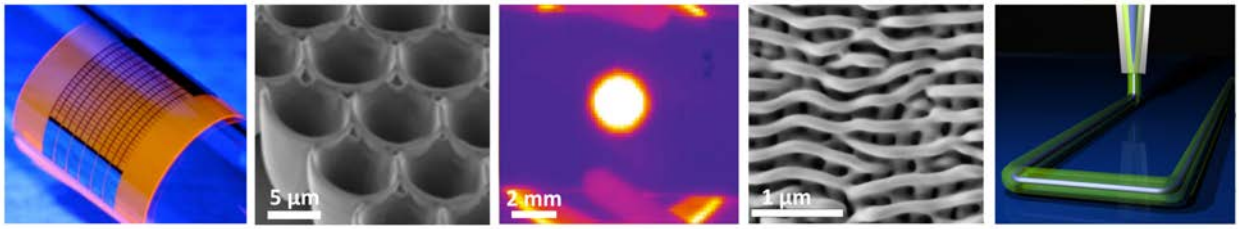
Research groups of the LMI-EFRC.

RG1 New Light Management Mechanisms: Establishing light management principles that challenge historical scientific ideas about conversion efficiency limits. RG1 is a theoretically motivated effort to develop building block components and optical mechanisms connecting to RG2, RG3, and RG4.

RG2 Solar Spectrum Control and Conversion: Investigating strategies for >40% photovoltaic conversion efficiency by exploiting spectrum splitting in novel architectures, photon recycling, and photon upconversion and downshifting in concentrating schemes.

RG3 Thermal Photon Harvesting: Implementing photonic design to develop new materials and structures for control of both the thermal emitter and the absorber for enhanced thermophotovoltaic performance.

RG4 Three-Dimensional Dynamic and Dispersive Materials: Developing three-dimensional dynamic and dispersive materials and synthesis methods that employ light-driven morphological evolution to achieve their own complex architectures in response to illumination conditions, and creating complex three-dimensional architectures using laser-assisted three-dimensional printing methods.



Representative LMI research efforts, from left: flexible downshifting luminescent solar concentrators; microscale parabolic concentrators for enhanced solar cell efficiency; infrared image of enhanced thermal emission via thermal extraction scheme; 3D structures grown by dynamic photolithography; light-enhanced 3D printing of concentric filaments

Light-Material Interactions in Energy Conversion (LMI)	
California Institute of Technology	Harry Atwater (Director), Andrei Faraon, Nate Lewis, Austin Minnich
Harvard University	Jennifer Lewis
Lawrence Berkeley National Laboratory	Paul Alivisatos, Eli Yablonovitch, Xiang Zhang
Stanford University	Mark Brongersma, Jennifer Dionne, Shanhui Fan
University of Illinois, Urbana-Champaign	Paul Braun, Ralph Nuzzo, John Rogers

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Energy Frontier Research in Extreme Environments (EFree)
EFRC Director: Russell J. Hemley
Lead Institution: Carnegie Institution of Washington
Start Date: August 2009

Mission: *To accelerate the discovery and synthesis of kinetically stabilized, energy-relevant materials using extreme conditions.*

The majority of our energy challenges are, at their core, due to intrinsic limitations of available materials with the required properties. By creating new materials with superlative properties including exceptional strength-to-weight ratios, resistance to corrosion, enhanced energy storage capacities, improved catalytic function and superior conversion and transport mechanisms, revolutionary energy savings can be achieved. Materials play a key role in all aspects of energy storage, conversion and utilization, and new materials are needed to improve performance in broad aspects of energy systems from solar, batteries, and fuels to new structural materials. In particular, entirely new strategies are required to access paradigm-shifting, transformative materials.

While traditionally most materials are created at or near ambient pressure, exploiting extreme environments, notably high pressures and temperatures, has tremendous potential for manipulating matter to synthesize next-generation materials with transformative impacts on energy. Moreover, materials in these extreme environments often exhibit optimal properties for energy applications. For example, extreme environments are capable of producing materials with unprecedented mechanical, thermal, and electronic properties.

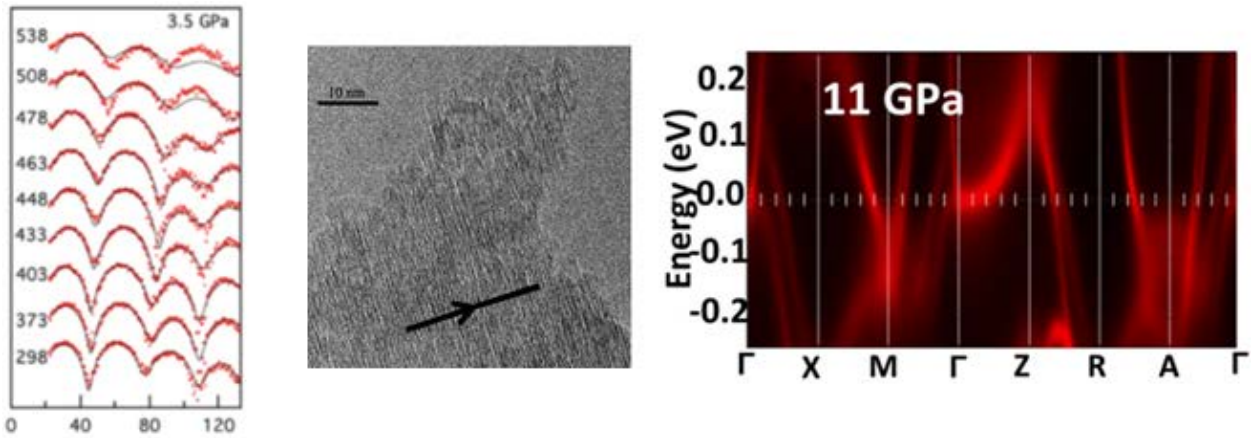

Figure 1 EFree employs a wide range of techniques for synthesizing and characterizing materials formed at extreme conditions. Left to right, high-pressure x-ray nuclear resonance spectroscopy provides measurements of the activation volume of polarons in battery materials such as $LiFePO_4$. Middle, TEM of our recently discovered sp3 carbon nanothreads shows periodic order transverse to the material. Right, advanced simulation methods, including DFT-DMFT, give detailed information on the evolution of spectral density of novel electronic materials such as FeSe.

EFree addresses specific knowledge gaps in the discovery of these paradigm-shifting, transformative energy materials. (Fig. 1). Novel materials to be discovered are broadly applicable to all classes of frontier energy research. The Center targets three focus areas, dictated by classes of energy materials and applications: advanced structural materials, novel energy conversion materials, and revolutionary energy transport materials. Our mission is to study materials under extreme conditions to deepen our understanding of materials behavior, enabling new materials to be discovered and, ultimately produced by design for practical use at ambient conditions.

To achieve this goal, EFree personnel work to synthesize fundamentally new materials and to exploit these for energy science through a focus on understanding kinetically stabilized routes to ambient pressure. We focus on studying, manipulating and ultimately controlling materials in regimes outside their fields of thermodynamic stability. In this, we employ new methodological approaches that allow access to new regions of phase space by treating recoverable high-pressure compounds as precursors for subsequent ambient-pressure manipulation. We also exploit pressure to mediate kinetically controlled synthesis of new materials in the solid state. Here, alternative reaction pathways are 'engineered' by using pressure to control the structure of precursors. In many systems, these pathways need not lead to thermodynamically stable reaction products, but to other new materials that can be stabilized by exploiting the kinetic limits of reaction rates. Finally, we also use chemical pressure and epitaxial growth to enhance the ambient-pressure stability of materials that exhibit exceptional high-pressure energy transport properties. By design, these approaches are highly synergistic and are exploited to forge effective collaboration across the Center using both experiment and theory (Fig. 2).

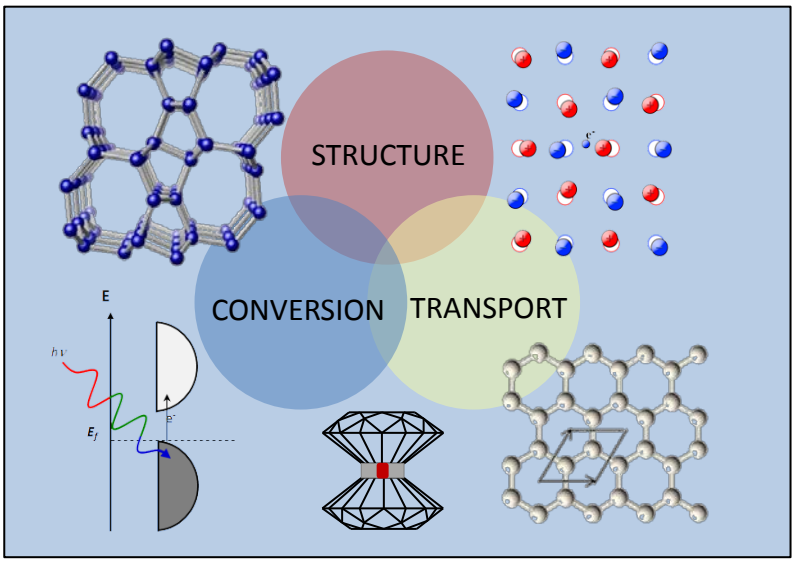

Figure 2 EFree uses of extreme conditions for synthesis and stabilization of advanced *structural, energy conversion, and transport* materials.

EFree thus employs fully integrated experiment and theory directed toward discovery, synthesis, and recovery of materials with exceptional properties using extreme conditions, especially high pressures and temperatures. The Center's overarching goal is in-depth understanding of kinetic stabilization mechanisms needed for the creation of revolutionary materials for robust real-world energy applications.

Energy Frontier Research in Extreme Environments (EFree)	
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California Institute of Technology	Brent Fultz
Colorado School of Mines	P. Craig Taylor
Cornell University	Roald Hoffmann
Lehigh University	Kai Landskron
Pennsylvania State University	John Badding, Vincent Crespi, Nasim Alem

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Catalysis Center for Energy Innovation (CCEI)
EFRC Director: Dionisios G. Vlachos
Lead Institution: University of Delaware
Start Date: August 2009

Mission Statement*: To focus on developing innovative, transformational heterogeneous catalytic technologies for utilization in future biorefineries to convert lignocellulosic (non-food-based) biomass into economically viable chemicals and fuels.*

The Catalysis Center for Energy Innovation (CCEI) focuses on developing innovative, science-based heterogeneous catalytic technologies for transformation of lignocellulosic (non-food-based) biomass materials into fuels and chemicals. Biomass offers a unique opportunity for a sustainable society with unprecedented impact on the US economy, energy security and independence. For this vision to be realized, we must overcome major scientific hurdles that are present due to the inherent complexity of biomass materials and associated processes.

The overall objectives of CCEI are to develop the enabling technologies for biomass conversion that lead to viable, economic operation of future biorefineries and to educate the workforce needed to further develop and implement these new technologies.

Since biomass feedstocks vary considerably by source and the number of candidate reactions is huge, the CCEI mainly focuses on developing a fundamental science base for controlling the scission and formation of C-H, O-H, C-C and C-O bonds by choosing a select number of reactions from a representative group of processes. Reactions typically take place in a complex, multiscale environment that renders the rational design of these processes and catalysts very challenging. Our overarching goal is to develop methods and concepts that form the foundations of modern biorefineries. The cornerstone of the CCEI lies in advancing catalysis and its integration into processes that deliver innovative technologies for the conversion of building blocks of cellulose and hemi-cellulose, i.e., of sugars (produced from the hydrolysis of biomass). We have assembled an interdisciplinary team of faculty and scientific staff that brings together the necessary expertise and synergism to tackle the challenges of these complex problems. We exploit various complementary catalytic technological platforms that are anticipated to play key roles in future biorefineries for energy, fuels, and chemical production. A unique trait of CCEI is the ability to carry out fundamental science in very complex problems, to introduce new inexpensive catalysts, and to integrate these catalysts into processes.

We have organized the following three crosscutting enablers as the backbone of the center's research to address the inherent complexities of the environment in which reactions take place: (1) synthesis of novel hierarchical multiscale materials with proper functional groups, (2) multiscale modeling, and (3) in situ characterization. These enablers are integrated within all research thrusts to quickly advance scientific breakthroughs. We develop multiscale models to understand the chemistry and to design new materials. We synthesize hierarchical multiscale materials that are hydrothermally stable, possess tunable porosity with bio-inspired functionality grafted active sites, and minimize resistance to molecular motion while allowing shape selectivity. Furthermore, we design cutting-edge characterization methods to probe reactions, often under in situ environment. Research at the Brookhaven National Laboratories is central for in situ characterization of the CCEI catalysts.

The CCEI outcomes include a fundamental understanding of the reaction mechanisms of representative biomass thermochemical transformations, a rational framework for multiscale hierarchical catalytic

materials and process design, science-based innovative technologies for biomass utilization, education of students and postdocs, and effective outreach/dissemination pathways to other scientists, students, and the public.

Catalysis Center for Energy Innovation (CCEI)	
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Brookhaven National Laboratory/Yeshiva University	Anatoly Frenkel
California Institute of Technology	Mark E. Davis
Columbia University/Brookhaven National Laboratory	Jingguang G. Chen
Georgia Institute of Technology	Christopher W. Jones
Lehigh University	Mark A. Snyder
Rutgers University	Marianthi G. Ierapetritou
University of Massachusetts Amherst	Wei Fan
University of Minnesota	Paul J. Dauenhauer J. Ilja Siepmann Michael Tsapatsis
University of Pennsylvania	Raymond J. Gorte Christopher B. Murray

Contact: Dionisios (Dion) G. Vlachos, CCEI Director, vlachos@udel.edu
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Center for Understanding and Control of Acid Gas-Induced Evolution of Materials for Energy (UNCAGE-ME)
EFRC Director: Krista S. Walton
Lead Institution: Georgia Institute of Technology
Start Date: August 2014

Mission Statement*: To develop a deep knowledge base in the characterization, prediction, and control of acid-gas interactions with a broad class of materials to accelerate materials discovery for large-scale energy applications.*

Acid gases are ubiquitous in multiple large-scale energy applications. For example, SO_x and NO_x are critical components of energy-related gas mixtures. Modern flue gas management involves catalysts that convert these species to less harmful products, as well as membrane and sorbents that can be degraded by these gases. As illustrated in Figure 1, degradation and deactivation effects of acid exposure can reduce access to active metal sites, block pores, reduce overall porosity, and destroy favorable textural properties. These degradation effects are often decisive factors in the practical use of materials such as sorbents for carbon capture, acid gas conversion, and natural gas purification. There are two major challenges in solving these problems: the mechanism of acid gas interactions with complex materials is poorly understood, and the ability to predict or prolong the lifetimes of functional materials is severely limited.

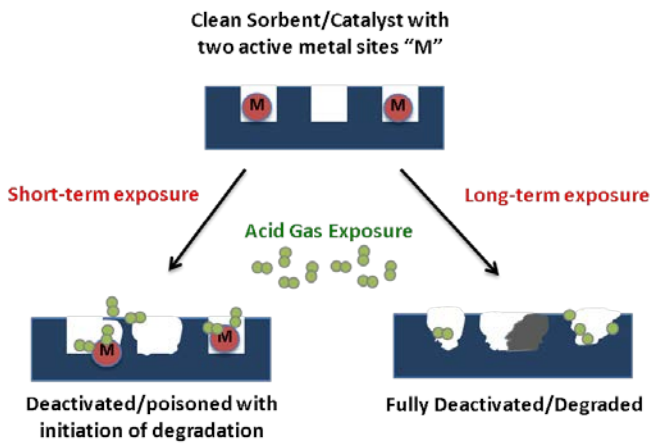

Figure 1: Changes induced in sorbents/catalysts upon short- and long-term exposure to acid gases. Materials are typically treated as passive in these environments, but can change dramatically upon exposure to SO_x, NO_x, and H_2S.

In this EFRC, we will seek to provide a fundamental understanding of acid gas interactions with solid materials through integrated studies of the interaction of key acid gases (CO_2, NO_2, NO, SO_2, H_2S) with a broad range of materials including 3D ordered structures such as metal-organic frameworks (MOFs), 3D disordered porous structures including carbide-derived carbons, supported amines, and oxides, and 2D analogues of these materials. We will combine the application of *in situ* molecular spectroscopic studies of both the surface functionalities and bulk structures of materials relevant to catalysis and separations under relevant environmental conditions with complimentary multiscale computational and theoretical modeling of acid gas interactions with solid matter. The Center will conduct research in four major thrust areas.

<u>Thrust I: Acid Gas Interactions with Model Oxide-Based Sorbents and Catalysts</u>

The interaction of small amounts of acid gases with sorbents/catalysts can dramatically affect their surface chemistry, useful lifetime and ease of regenerability. The overarching goal of Thrust I is to gain a molecular level understanding of the surface chemistry of acid gas interaction with model sorbents (oxides) and catalysts (supported metals and oxides) and their structural evolution. This will provide structure-performance relationships for the rational design of more efficient sorbents for acid gas removal and more stable catalysts for CO_2 utilization.

Thrust II: Effect of defects and dopants on acid gas interactions with ordered porous materials

The stability of crystalline, high surface area adsorbents such as metal-organic frameworks (MOFs) towards acid gases is a major obstacle in their development as successful materials for energy-related applications. The main goal of this Thrust is to determine the structural features that control this (in)stability. The main hypothesis of this Thrust is that acid gas stability of such materials, and their potential degradation upon exposure to acid gases, are driven by local defects or centers susceptible to acid attack distributed throughout the material. If this is the case, characterization and control of the local defects/reactive centers is vital to enhancing material performance.

Thrust III: Interaction of acid gases with disordered porous solids

A core aim of the Center is to obtain fundamental information about acid gas interactions that are applicable to broad classes of materials. Our efforts using crystallographically ordered porous materials in **Thrust II** will be complemented by **Thrust III**, which will center on disordered porous materials including templated or carbide-derived carbons and supported amines. Both classes of materials have significant heterogeneity with regard to structure in the final form used in acid gas separations and catalysis, yet both are made from more well-defined precursors or supports, which gives a basis for understanding the complex structures of these materials. **Thrust III** aims to create a body of knowledge that ultimately relates the support structure and amines/heteroatoms/defects to its interaction, stability and/or degradation characteristics in the presence of NO_x, SO_x, CO_2, and H_2O.

Thrust IV: External Surfaces of Porous Materials

The work proposed in Thrusts II and III assumes that degradation of porous materials is likely to take place through chemical reactivity associated with defects. Thus, in those Thrusts, defects *inside* a material's pores are the focus of attention. In Thrust IV, we focus attention on another unavoidable class of defects present in any real porous material – *external surfaces*. A key goal of this Thrust is to characterize the surfaces of representative nanoporous materials and to understand how these surfaces differ from the pore structure defined by the "bulk" material. Demonstrating methods to achieve this goal will create many opportunities for targeted synthetic and post-synthetic strategies to enhance material performance. Understanding the surface structure of nanoporous materials will be beneficial not only in controlling interactions of these materials with acid gases but also in other important applications.

Center for Understanding and Control of Acid Gas-Induced Evolution of Materials for Energy (UNCAGE-ME)	
Georgia Institute of Technology	Krista Walton, Christopher Jones, David Sholl, Sankar Nair, Michael Filler, Ryan Lively, Thomas Orlando
Oak Ridge National Laboratory	Sheng Dai, Gernot Rother, Tjerk Straatsma, Bobby Sumpter, Zili Wu
University of Alabama	David Dixon
University of Florida	Susan Sinnott
Lehigh University	Israel Wachs
Washington University St. Louis	Sophia Hayes
University of Wisconsin	JR Schmidt

Contact: Krista Walton, Associate Professor and Marvin R. McClatchey and Ruth McClatchey Cline Faculty Fellow, School of Chemical & Biomolecular Engineering, Georgia Tech, walton@efrc.gatech.edu 404-894-5254, http://efrc.gatech.edu

Integrated Mesoscale Architectures for Sustainable Catalysis (IMASC)
EFRC Director: Cynthia Friend
Lead Institution: Harvard University
Start Date: August 2014

Mission Statement: *To develop a fundamental understanding of how to design and use novel mesoporous catalyst architectures for sustainable conversion and production of platform chemicals through selective oxidation and selective hydrogenation.*

IMASC seeks fundamental understanding for the purpose of developing novel mesoporous catalysts for sustainable processes that drive the conversion and production of platform chemicals. Understanding reaction kinetics, building innovative catalyst architectures and improving reaction selectivity under catalytic conditions will lead to new opportunities to decrease fuel consumption through creation of more efficient processes. IMASC will advance the scientific grand challenge question "how do we design and perfect atom- and energy-efficient synthesis of revolutionary new forms of matter with tailored properties?" through integrated efforts at Harvard University, Tufts University, Lawrence Berkeley and Lawrence Livermore National Laboratories, University of Notre Dame and the Fritz Haber Institute.

Objectives:

- To understand how to control the selectivity of complex, coupled reactions for important chemical processes, including selective oxidation and selective hydrogenation.
- To synthesize novel porous metal alloy and metal oxide catalysts with specific active sites for activation of key reactants
- To understand and control surface composition and defect structure to optimize catalytic performance informed by advanced imaging, X-ray spectroscopy and theoretical tools.

IMASC Research Team and Research Focus Areas:

The IMASC team includes chemists, physicists, chemical engineers, and materials scientists who will perform research organized into three research Focus Areas: (1) Reaction Mechanisms and Modeling of Active Sites; (2) Understanding Performance under Catalytic Conditions; and, (3) Structure, Composition and Electronic Properties (cross-cutting theme).

Focus Area 1 (FA1): ***Reaction Mechanisms and Modeling of Active Sites.*** The mechanisms of selective catalytic hydrogenation and oxidation will be investigated on the molecular scale on well-defined single crystal surfaces and on complex mesoporous materials under highly controlled conditions (Figure 1). Studies will lead to a comprehensive model of reaction mechanisms, including elementary chemical steps and the nature of active sites. Investigations will include of a range of (host) materials, including Cu, Ag, Au and metal oxides (TiO_2, CeO_2, ZnO) combined with active metals (e.g., Pd, Ni and Pt). Designed meso- and nanoporous materials will also be synthesized as catalysts. Advanced theoretical methods will play a

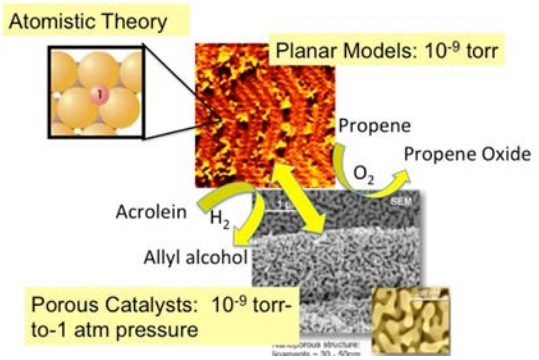

Figure 1: Illustration of IMASC research using planar models and nanoporous catalyst materials in conjunction with advanced theory to understand selective oxidation and selective hydrogenation.

guiding role in understanding this experimental work. Specific investigations are: (1) selective hydrogenation of unsaturated carbonyl compounds to unsaturated alcohols by alloys of Cu, Ag and Au with minor amounts of Ni and Pd; and (2) selective oxidation of olefins over Ag or Au alloyed with Pd or hybridized with metal oxides. FA1 will focus on relating reaction selectivity to materials composition and structure as we build up a sophisticated general model for these classes of reactions. Atomistic theory will provide insight into the interplay of electronic and geometric structure in determining bonding and reactivity of molecular intermediates. This research will inform catalyst design through close coupling with research in the other Focus Areas.

Focus Area 2 (FA2): Understanding Performance under Catalytic Conditions. The efficacy of specific mesoporous architectures for catalytic activity and selectivity under realistic reaction conditions will be established through reactor studies. The unique capabilities of temporal analysis of products (TAP) experiments to quantitatively determine rate constants will be combined with studies in flow reactors for vapor phase processes and batch reactors for liquid phase reactions. *In situ* (1 atm. pressure and liquid phase) This research will be closely coupled with the research in FA1 to refine the working hypotheses about what materials properties and reaction conditions yield better performance (Fig. 1). Close coupling with FA 3 will be used to model the effect of mesoporous geometry to the local surface concentrations of reactants and products through multi-scale simulations.

Focus Area 3 (FA3): Understanding Performance under Catalytic Conditions; and, (3) Structure, Composition and Electronic Properties. This cross-cutting research theme, will both develop and use advanced theoretical and experimental tools to understand the effects of surface composition, structure and electronic properties on the catalytic reactivity of the model and mesostructured materials and the scale-up from the atomic scale to the mesoscale architectures. Powerful microscopies and x-ray probes will be combined to understand the working reactive surfaces. Multiscale theory will provide a fundamental understanding of both the materials synthesis and the evolution of the mesoscale materials under reactive conditions (Figure 2).

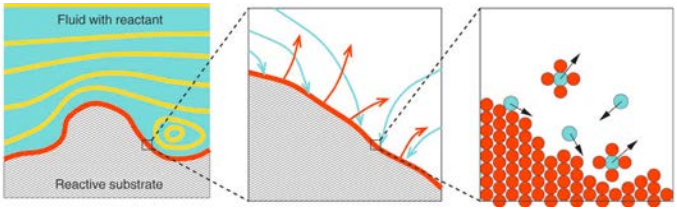

Figure 2: Illustration of the multiscale approach based on for capturing the transport of reactants and products at various scales, from the macroscopic (left) to mesoscopic (middle panel, fluid flow and reactant/product concentrations modeled separately), to the molecular level (right).

Integrated Mesoscale Architectures for Sustainable Catalysis (IMASC)	
Harvard University	Cynthia Friend (Director), Efthimios Kaxiras (Deputy Director), David C. Bell, Jennifer Hoffman, Robert Madix
Tufts University	Maria Flytzani-Stephanopoulos
Lawrence Berkeley National Laboratory	Miquel Salmeron
Lawrence Livermore National Laboratory	Juergen Biener
University of Notre Dame	Franklin (Feng) Tao
Fritz Haber Institute	Alexandre Tkatchenko

Contact: Cynthia Friend, Director, cfriend@seas.harvard.edu
617 495 4052

Center for Geologic Storage of CO_2 (GSCO2)
EFRC Director: Robert J. Finley
Lead Institution: University of Illinois at Urbana-Champaign
Start Date: August 2014

Mission Statement*: To generate new conceptual, mathematical, and numerical models applicable to geologic storage systems in specific and strategically identified research areas, based on uncertainty and limitations observed in field pilots and CO_2 injection demonstration projects, laboratory experiments, and the experience of researchers in industry-sponsored applied research.*

The Center for Geologic Storage of CO_2 aims to improve fundamental understanding of the storage reservoir in terms of two of the most important questions regarding the commercial storage of CO_2:

- How will the injected CO_2 be distributed within the storage reservoir and how can we improve predictions of its location?
- What is the mechanism of injection-induced microseismicity, and can we control and predict its occurrence?

To answer these questions, geology, geophysics, geomechanics, and multiphysics flow and transport are the four areas of research. The four-year scientific research goals of the Center are to:

- quantify and characterize the relative impact of potentially significant advances or weaknesses in commercial-scale CO_2 storage technology in order to prioritize and direct the research of the Center;
- use fundamental and basic scientific principles as the basis for investigating existing storage technology gaps, with the objective of defining usable, near-term solutions;
- employ pore- and molecular-scale simulations that are effectively and properly scaled to the nano, micro, and macro scale of laboratory data, and mega scale of injection site and basin data; and
- ensure that new, reliable methods derived from basic science studies are field ready and compatible for industry application in CO_2 geologic storage projects.

Geology: Characterization and Geocellular Modeling: Understanding subsurface flow dynamics, including CO_2 plume migration and trapping, requires understanding a diverse set of geologic properties of both the reservoir and caprock, from the pore scale to the basin scale. The uncertainty in site-specific geology stems from the inherent variation in rock types, depositional environments, and reservoir properties. Methodologies and tools are needed to represent reservoir properties from the pore to field scale and take into consideration small-scale features that could significantly affect storage integrity. Geologic attributes affecting reservoir behavior must be characterized. For example, the amount of CO_2 trapped in the reservoir by capillary trapping processes is a complicated nonlinear function of the spatial distribution of permeability, permeability anisotropy, capillary pressure, relative permeability of brine and CO_2, permeability hysteresis, and residual gas saturation. Importantly, the spatial variability (heterogeneity) of these petrophysical attributes that control capillary trapping is defined by a multiscaled and hierarchical sedimentary architecture within sandstone formations.

Geophysics: The application of traditional approaches to interpret surface-based seismic data to detect injected CO_2 may be limited as a result of "cultural noise" local to a site or highly complex near-surface conditions. Because of the latter possibility, we are including a vertical seismic profile (VSP) seismic effort in our research strategy. Cultural noise and near-surface conditions do not typically affect seismic data acquired with deep sensors, yet VSP data have shown evidence of CO_2 plumes only with some degree of uncertainty. A reason for the uncertainty in detecting injected CO_2 with VSP data is that only the P-wave mode has been used in VSP studies. Therefore, the use of both P- and S-wave modes in the VSP analyses is expected to better illuminate the structure of the reservoir and determine if a

combination of P- and S-wave data significantly improve imaging the CO_2 plume. Improved geophysical interpretation will provide important data to calibrate flow models and improve regulatory and public confidence to track CO_2 in the reservoir.

Geomechanics: Improved understanding of coupled stress, strain, and flow response to injection-induced pressure transients at all scales will be investigated. Laboratory experiments will discover the mechanism(s) accountable for potential changes to porous media given specific externally applied stresses and injection conditions (e.g., slow slipping deformation, grain crushing, microseismic events). Furthermore, laboratory experiments will illustrate which attribute(s) trigger microseismicity, and the results will be related to field-scale observations of microseismicity. Detailed characterization of the type of microseismicity (e.g., source mechanism, stress drops, ratio between radiated seismic energy to seismic moment, spatial-temporal distribution of b-values) will provide constraints on the CO_2 plume's extent, pressure, and stress front. This new knowledge will assist in risk mitigation related to the triggering potential of felt earthquakes and in dealing with public acceptance.

Multiphysics Flow and Transport: Subtle small-scale heterogeneities can influence CO_2 migration. At an even smaller scaller, the displacement of reservoir brine by injected CO_2 is highly dependent upon pore space morphology and interconnectivity, and the complex interactions between viscous and capillary forces which change with fluid pressure and temperature as well as wettability properties of the rock. These fundamental properties of CO_2 in its unique state as a supercritical fluid confined with brine within a porous media depend upon molecular-scale phenomena that will be evaluated using molecular dynamics simulation. These include interfacial tension, density, viscosity, and contact angle with reservoir minerals, all as a funciton of reservoir temperature and pressure. The results will be used to inform and parameterize pore and continuum scale model development, including selection of appropriate equations of state. Pore scale models will be tested against pore scale experiments of CO_2 injection, where CO_2 flow paths and pressure are evaluated as a function of pore scale geometry, wettability variation, and surface mineral reactions. Constitutive relationships will be developed from pore scale experiments and modeling, and used in continuum scale modeling that will be informed by simultaneous capillary pressure – saturation and CO_2 flooding experiments. The improved continuum-scale models will be used to predict field performance of CO_2 injection, with the goal of more accurately predicting CO_2 saturation and presssure distribution within CO_2 storage reservoirs.

Center for Geologic Storage of CO_2 (GSCO2)	
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University of Texas-Austin	Robert Hardage, Charles Werth, Michael DeAngelo, Diana Sava
NORSAR	Volker Oye, Bettina Goertz-Allmann
Wright State University	Robert Ritzi, Naum Gershenzon
Schlumberger	Sergey Stanchits, Donald Lee, Robert Will
University of Southern California	Muhammad Sahimi, Theodore Tsotsis, Kristian Jessen
SINTEF	Michael Jordan, Erik Lindeberg, Erling Fjaer
University of Notre Dame	Kenneth Christensen
National Energy Technology Laboratory	Angela Goodman

Contact: Robert J. Finley, GSCO2 Director, finley@illinois.edu
217-244-8389

Center for Nanoscale Control of Geologic CO_2 (NCGC)
EFRC Director: Donald J. DePaolo
Lead Institution: Lawrence Berkeley National Laboratory
Start Date: August 2009

Mission Statement*: To enhance the performance and predictability of subsurface storage systems by understanding the molecular and nanoscale origins of CO_2 trapping processes, and developing computational tools to translate to larger-scale systems.*

The vision for the Center is to understand, predict, and enhance the performance of underground CO_2 storage systems. Specific goals are to produce (1) a next-generation understanding of the nanoscale-to-mesoscale chemical-mechanical behavior of shale - a critical material for a low-carbon energy future, (2) quantitative models for the efficiency of reservoir capillary trapping and its effect on solution and mineral trapping, (3) methods to predict mineralogical trapping, and (4) theory, experimental data, and computational tools to allow nanoscale effects to be translated to mesoscale and continuum scale model equations and parameters.

The NCGC consists of a team of highly qualified investigators with expertise in, and access to, the most advanced analytical and computing facilities available for furthering fundamental knowledge of the geochemical aspects of geologic carbon storage, in particular the chemistry of mineral-fluid interfaces and fluid-fluid interfaces that control the physics of fluid flow and chemical reactions. Experimental investigations will transition from simplified analogue materials to more realistic geologic materials. Newly developed characterization and experimental approaches will be integrated with mesoscale chemical-mechanical-hydrologic modeling and simulation to achieve a transformational predictive capability for stratigraphic- and reservoir CO_2 trapping efficiency and reliability.

Scientific gaps that the Center will address are:

1. The origin and evolution of wetting properties of complex, reactive fluids in contact with common minerals encountered in carbon storage
2. Reactivity between fluids and minerals in nanoporous to macroporous rocks
3. Long term-evolution of capillary-trapped CO_2
4. The response of fractured shale to intrusion of CO_2-containing mixed fluids
5. Theory and computational tools that allow the large scale and long-timescale evolution of fluid-rock systems to be simulated such that they reflect the nanoscale and mesoscale properties of geological materials at far-from equilibrium conditions.

The research of the NCGC is divided into three Thrust Areas that address (1) the sealing effectiveness of fractured shales, (2) reservoir processes that control secondary trapping (capillary, dissolution and mineral trapping) and (3) developing the computational tools and insight necessary to model mesoscale couplings and material properties and dynamics. Systems of study will include well-characterized natural rock and mineral samples, and synthetic materials fabricated by established methods and methods to be developed. A key aspect of our approach is to bring multiple characterization methods, and diverse complementary expertise, to bear on the same experiments, and to integrate modeling and simulation with experiments. The Center will leverage the characterization and computational facilities at LBNL (Advanced Light Source, Molecular Foundry, National Energy Research Scientific Computing Center), ORNL (Spallation Neutron Source, High Flux Isotope Reactor, Center for Nanomaterials Science) and other synchrotron facilities (Advanced Photon Source at Argonne Laboratory, Stanford Synchrotron Radiation Laboratory, and the National Synchrotron Light Source at Brookhaven Lab).

Stratigraphic Trapping by Fractured Shale: Thrust Area 1 aims to provide an understanding of the nanometer to meter scale properties of seal rocks and fractures. The research plan is structured around four phenomena: the geochemistry of confined fluids (Task 1.1), mineral reactivity and colloid generation at fracture surfaces (Task 1.2), coupled mass transfer and mineral reactivity at the boundary between the fracture and the rock matrix (Task 1.3), and coupled multiphase flow, geochemistry, and geomechanics in the fracture itself (Task 1.4). The fifth phenomenon, the role of organics, is cross-cutting and will be examined in each task.

Reservoir Trapping Processes: Thrust Area 2 aims to provide a better understanding of storage security beyond the stratigraphic seal; to quantify the residual and mineral trapping of CO_2 in GCS reservoirs. The research plan is structured around 4 tasks, investigating physicochemical controls on residual trapping (Task 2.1), residual trapping estimates at short times (Task 2.2), the long term stability of residual storage (Task 2.3), and mineral trapping processes (Task 2.4).

Modeling Approaches for the Mesoscale Challenge: Thrust Area 3 aims to build on the progress made in NCGC to address the mesoscale challenge from the DOE BES Basic Research Needs report *From Quanta to the Continuum: Opportunities for Mesoscale Science.* This Thrust is organized into three broader topics on 1) Mesoscale Reaction Rates: Upscaling Reaction Rates from Molecular to Pore Scale, 2) Mesoscale Fractures: Modeling Fracture Networks, and 3) Mesoscale Reactive Transport: Theoretical and Computational Approaches for Upscaling to the Mesoscale.

Center for Nanoscale Control of Geologic CO_2 (NCGC)	
Lawrence Berkeley National Laboratory	Don DePaolo (Director), Carl Steefel (Deputy Director, Theme Lead), Jonathan Ajo-Franklin (Theme Lead), Ian Bourg (Theme Lead) PARTICIPANTS: Benjamin Gilbert, Tim Kneafsey, Sergi Molins-Rafa, Peter Schuck, Tetsu Tokunaga, David Trebotich, Jiamin Wan
Oak Ridge National Laboratory	Andrew Stack, Gernot Rother, Larry Anovitz
Ohio State University	David Cole
Purdue University	Laura Pyrak-Nolte
Stanford University	Sally Benson, Hamdi Tchelepi
Washington University, St. Louis	Young-Shin Jun

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510-486-7560, http://esd.lbl.gov/research/facilities/cncgc/

Center for Advanced Solar Photophysics (CASP)
EFRC Director: Victor I. Klimov
Lead Institution: Los Alamos National Laboratory
Start Date: August 2009

Mission Statement*: To exploit fundamental interactions between nanomaterials and light with the goal of producing disruptive advances in the efficiency of solar energy conversion.*

Solar light is a tremendous resource of clean energy. Widespread use of solar energy will require technologies that offer a dramatic increase in photoconversion efficiency *simultaneously* with reduced cost, which will require disruptive advances relying on new physical principles and novel materials. Quantum-confined semiconductor nanocrystals (NCs) are fascinating structures offering tremendous promise in the area of solar energy conversion into either electricity or chemical fuels. The development of NC-based approaches lies at the intersection of chemistry, physics, materials science and engineering. The Center for Advanced Solar Photophysics (CASP) unites world-class experts spanning these disciplines toward the purpose of harnessing the unique properties of quantum-confined semiconductor materials to facilitate the realization of the next generation of low-cost, high-efficiency solar photoconversion systems.

As an established leader in the design, synthesis and exploitation of novel nanomaterials that exhibit advanced, solar-relevant physical phenomena, CASP continually strives to achieve new levels of efficiency in NC-based photovoltaic (PV) devices. In addition, we are vigorously investigating a broad range of approaches toward applying the unique properties of nanomaterials to boost the efficiencies of existing or emerging PVs above present limits. This multifaceted research program is captured in four main Themes (Fig. 1), each of which strives to address a specific, essential question:

- *How can we enhance the efficiency of carrier multiplication in engineered nanomaterials?* The observation of carrier multiplication (the generation of multiple excitons by a single absorbed photon) in NC-based PVs is a singular example of advanced nanoscale physics being brought to bear on real solar cells. While the efficiencies of this phenomenon in NCs are enhanced relative to those in bulk semiconductors, they are still not high enough to considerably impact power conversion efficiency limits. In the **Carrier Multiplication by Design Theme**, we investigate how structure-based control over carrier cooling, and carrier-photon and carrier-carrier interactions can drive CM efficiency to the theoretical limit.
- *How can we use nanomaterials to harness the full solar spectrum more efficiently?* The ultimate conversion efficiency of standard PV devices is limited by two main channels of energy loss: the inability to capture lower-energy, sub-band-gap photons, and the dissipation of the excess energy of above-band-gap photons as heat. In the **Advanced Photon Management Theme**, we employ NCs to make better use of such photons through up- or down-conversion schemes (collectively called "spectral reshaping") as well as to augment sunlight harvesting with inexpensive luminescent solar concentrators.
- *How can we manipulate inter-particle coupling to control charge and energy transport in mesoscopic assemblies?* Our understanding of the unique physical phenomena that arise in nanoscale materials has rapidly advanced, but to be relevant, these phenomena must be active in macroscopic devices. In the **Functional Mesoscopic Assemblies Theme**, we develop approaches for integrating engineered NCs into mesoscopic assemblies that retain original functionalities and augment them with efficient carrier transport and other emergent phenomena associated with controlled inter-particle coupling.

- *How can unique nanoscale phenomena be exploited in advanced photoconversion devices?* Currently, optimizing the performance of solar cells based on NCs relies on tuning their properties to meet the needs of a conventional PV architecture. To overcome the limitations of this traditional concept, in the **Advanced Device Architectures Theme**, we will explore new photoconversion schemes tailored for a specific nanostructure and optimized *via* convergent evolution of both the material and the device architecture.

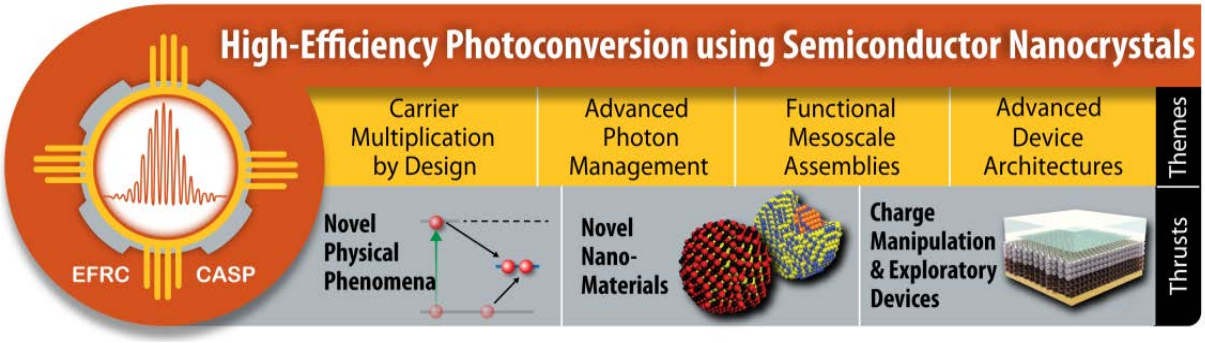

Figure 1. Research themes address essential questions using the capabilities housed within CASP's three Thrusts.

To tackle these research Themes we utilize world-class capabilities and expertise organized within a Thrust structure that supports a comprehensive approach to all relevant aspects of each problem. The **Novel Physical Phenomena (NPP) Thrust** combines capabilities in optical and scanning-probe spectroscopies emphasizing state-of-the-art femtosecond and single-NC techniques with theory, modeling and simulation that encompass effective-mass, tight-binding, quantum chemistry, and atomistic techniques. The **Novel NanoMaterials (NNM) Thrust** provides advanced capabilities for implementing materials-by-design concepts in the synthesis of engineered NCs and multifunctional nanocomposites using colloidal and plasma-based techniques. Finally, the **Charge Manipulation and Exploratory Devices (CMD) Thrust** houses unique capabilities for probing transport in mesoscopic QD arrays, as well as for fabrication and characterization of NC-based devices with architectures that exploit novel nanoscale physics.

Center for Advanced Solar Photophysics (CASP)	
Los Alamos National Laboratory	Victor Klimov (Director), Jeff Pietryga, István Robel, Kirill Velizhanin
National Renewable Energy Laboratory	Matt Beard (Associate Director), Justin Johnson, Joey Luther, Art Nozik
University of California, Irvine	Matt Law
University of Minnesota	Uwe Kortshagen
George Mason University	Alexander Efros, Andrew Shabaev
University of Pennsylvania	Cherie Kagan
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Nanostructures for Electrical Energy Storage (NEES)
EFRC Director: Gary W. Rubloff
Lead Institution: University of Maryland
Start Date: August 2009

Mission Statement*: To reveal scientific insights and design principles that enable a next-generation electrical energy storage technology based on dense mesoscale architectures of multifunctional nanostructures.*

In its first phase (2009-2014) NEES focused on the design and electrochemistry of heterogeneous, multifunctional nanostructures as the building block for electrical energy storage (EES) configurations offering high power and stability during charge/discharge cycling. Using prototype materials relevant to Li ion batteries, NEES has employed novel single-nanostructure platforms and "forests" of densely packed nanostructure arrays to fuel insights into nanostructure electrochemistry. Along the way, fundamental studies of ion transport, elucidation of electron vs ion transport kinetics, and experience with dense nanoelectrode arrays converged to create a growing focus on regular and random 3-D architectures of nanostructures and important consequences which follow.

In its second phase NEES is concentrating on the arrangement of precision nanostructures into mesoscale architectures and the new scientific challenges that result. These investigations begin with

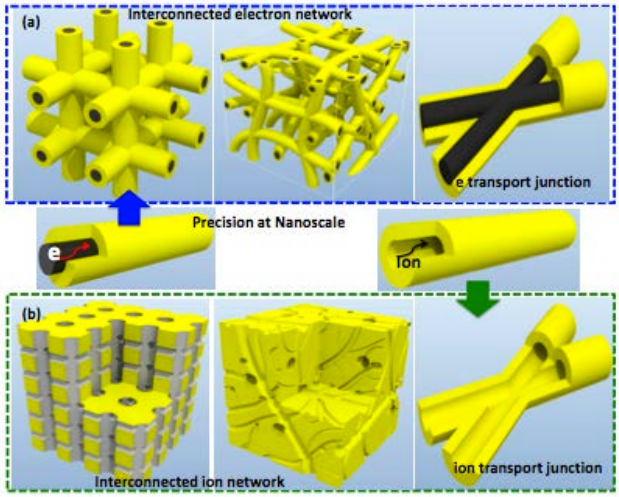

Figure 1. Precision nanostructures arranged into mesoscale architectures.

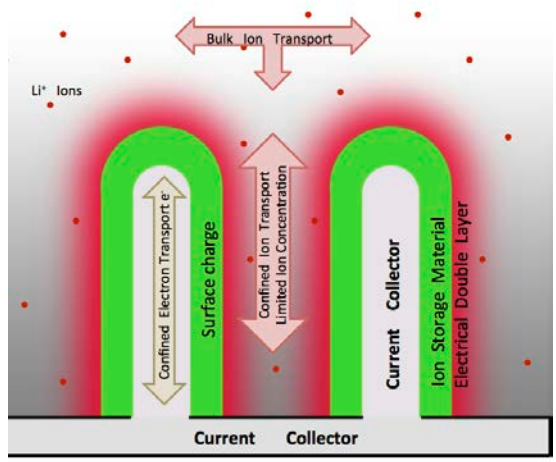

Figure 2. Ionics and electrodics phenomena in dense nanostructure forests.

achievable architectures (Fig. 1), which raise questions such as: What 3-D synthesis strategies translate the benefits of precision nanostructures to dense mesoscale assemblies? How does the EES behavior of regular/ordered architectures compare with that of pseudorandom counterparts? How well do metrics for the latter, such as porosity and tortuosity, forecast EES performance? The dense packing of nanostructures into mesoscale architectures in turn creates new scientific concerns of ion and electron transport (Fig. 2), inviting such fundamental questions as: Will electrolyte nanoenvironments contain and supply ions in sufficient concentration to fully utilize EES storage layers in the nanostructures? What design guidelines are needed to balance electron and ion transport (in current collectors and electrolyte respectively) to achieve high power? Will surface charge vary along nanostructures, and what role might it play in electrolyte ion transport phenomena (e.g. electrokinetics)?

NEES will also pursue the challenge of understanding capacity degradation and failure in nanostructure-based EES - an ambitious effort to establish a science of nanostructure degradation, simultaneous with or in advance of a prototype nanostructure-based EES technology. NEES will employ advanced nano/micro scale platforms (Fig. 3) to identify degradation/failure in both early (<10's of cycles) and extended-term (~50-1000 cycles), from which specific follow-on test structures can be constructed and tested to validate proposed degradation mechanisms.

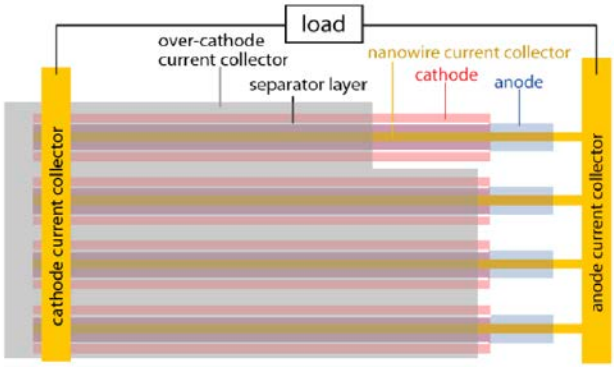

Figure 3. Horizontal nanobatteries as degradation test sites.

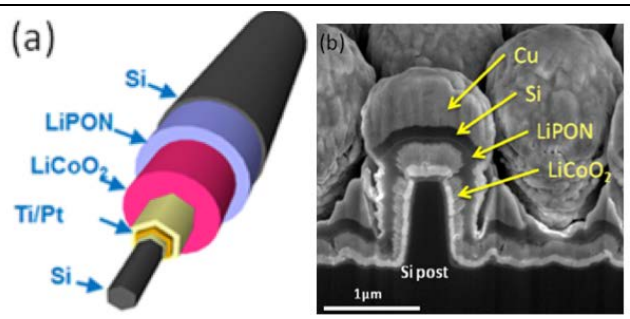

Figure 4. 3-D solid state nanobatteries.

As an ambitious alternative to safe, high performance EES, NEES will expands its foray into the synthesis of solid state electrolytes and interdigitated 3-D solid state batteries (Fig. 4) to reduce degradation, increase safety, and maximize active mass and volume. Solid state electrolytes are also critical for proposed EES as passivation/protection layers for metal anodes.

In pursuing dense architectures of precision multifunctional nanostructures and their profound implications for mesoscale science, NEES will employ research platforms based on conventional and advanced Li ion battery systems. In addition, NEES has initiated and will research selected beyond-Li-ion battery chemistries, including multivalent systems (e.g., Mg^{2+}), metal anode (e.g., Li-O_2), and solid state configurations, particularly where NEES' unique capabilities in precision nanostructures, synthesis and characterization, and modeling are poised to address and advance key scientific questions in these domains.

Nanostructures for Electrical Energy Storage (NEES)	
University of Maryland	Gary W. Rubloff (Director), Sang Bok Lee (Deputy Director), John Cumings, Reza Ghodssi, Chunsheng Wang, YuHuang Wang, Liangbing Hu, Janice Reutt-Robey, Bryan Eichhorn, Elizabeth Lathrop (Assoc. Director for Programs)
Sandia National Laboratories	John Sullivan (Assoc. Director fro Sandia), Kevin Leung, A. Alec Talin, Katherine Jungjohann, C. Tom Harris
University of California, Irvine	Reginald Penner, Zuzanna Siwy, Phil Collins
Yale University	Mark Reed
University of Florida	Charles R. Martin
University of California, Los Angeles	Bruce Dunn
University of Utah	Henry White
Michigan State University	Yue Qi

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Center for Excitonics (CE)
EFRC Director: Marc Baldo
Lead Institution: Massachusetts Institute of Technology
Start Date: August 2009

Mission Statement*: To supersede traditional electronics with devices that use excitonics to mediate the flow of energy.*

Exciton - *a quasiparticle excitation consisting of a bound electron and hole that mediates the absorption and emission of light, especially in disordered and low-dimensional materials.*

In this Center, we seek to supersede traditional electronics with devices that use excitons to mediate the flow of energy. Mastering the properties of excitons offers the ability to guide energy at the nanoscale, and transform it with a flexibility that is impossible in conventional systems. For example, within the center we have exploited the flexibility of excitonic transformations, using singlet exciton fission to generate multiple carriers in a solar cell at an efficiency of > 1.26 electrons per photon, exceeding the unity limit in conventional electronic materials.

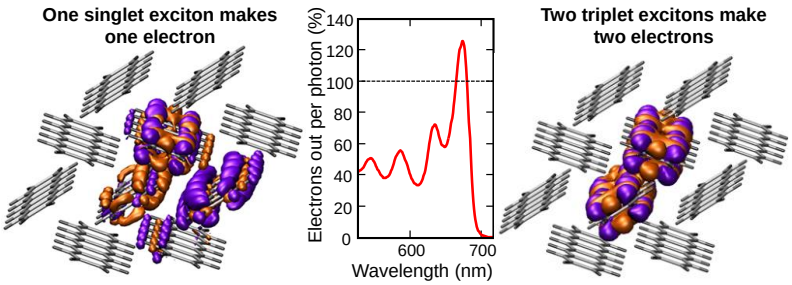

Figure 1. Unlike electrons, excitons can be split. In the Center for Excitonics, we have used singlet exciton fission in pentacene to generate at least 1.26 electrons per photon.

Team 1: Multiple Carrier Excitonics for Solar Cells
(*Baldo, Bawendi, Dauler, Jarillo-Herrero, Kong, Levitov, Tisdale, van Voorhis*)

Singlet exciton fission is one focus of our fundamental studies of exciton splitting in different materials. In conventional nanocrystalline semiconductors, *Bawendi* and *Dauler* have performed extensive single molecule spectroscopy showing that excitation of secondary electrons must out-compete rapid thermalization losses. But in molecules *Baldo* and *Van Voorhis* demonstrated that these losses are spin disallowed, meaning that exciton fission can be almost perfectly efficient. Graphene is a third, and special case. Prior work by *Jarillo-Herrero* and *Levitov* demonstrated that graphene may be capable of generating ~ 10 carriers per photon if the hot carriers can be extracted in time.

This team's continuing focus is *dynamics in states of multiple excitations*. We will pursue three collaborative projects that build on our prior work: (i & ii) multiexcitons and charging in semiconductor nanocrystals, and (iii) sensitized triplet transfer for singlet exciton fission. We also introduce a new project: (iv) hot carriers in graphene.

Team 2: High Density Excitonics for Solid State Lighting
(*Bawendi, Baldo, Bulovic, Englund, Gradecak, Jarillo-Herrero, Kong, Stach, Tisdale, van Voorhis*)

In Team 1, we seek higher solar cell efficiencies by exploiting unique properties like exciton fission. The focus of Team 2 is the second general advantage of excitonic materials: *ease of fabrication.* Given the dramatic reduction in manufacturing cost compared to conventional devices, it is notable that organic and quantum dot-based LEDs exhibit efficiencies just as large as conventional, crystalline LED materials.

The catch is that organic and quantum dot-based LEDs cannot be driven to high brightness like conventional LEDs. *This is the problem of 'high density excitonics'.* This team will pursue three approaches to high brightness LEDs. *Baldo, Bulovic* and *Van Voorhis* will work on OLEDs with short exciton lifetimes. *Bulovic, Bawendi,* and *Tisdale* will reduce losses in QD-LEDs, and *Jarillo-Herrero* and *Kong* will work with the other LED groups to introduce a new class of LEDs based on 2d materials, notably WSe_2. We will also use a wide-field, spin-based technique developed by *Englund* to map excitons and charges in disordered materials. Several complementary non-destructive spectroscopic and structural characterization techniques developed by *Gradecak* with nano- and atomic-scale resolution will be used to directly correlate structure and optical properties of disordered materials. In collaboration with BNL (*Stach*), these results will be compared with the state of the art commercial semiconductor LED structures.

Team 3: Quantum Excitonic Transport
(*Aspuru-Guzik, Nelson, Baldo, Black, Dinca, Levitov, Jarillo-Herrero*)

Photosynthesis is the best example of the benefits of excitons in a practical, nanostructured system. Like our devices, photosynthesis exploits excitons and exhibits much larger levels of long range disorder than conventional solar cells. Indeed, this team was partly motivated by recent ultrafast experiments on the excitonic properties of photosynthetic complexes. To aid in the interpretation of these measurements, the *Aspuru-Guzik* group proposed an experimental protocol to systematically characterize both coherent and dissipative processes at the level of the evolving quantum state of the probed chromophores. In collaboration with postdoc *Joel Yuen-Zhou*, and the *Bawendi* and *Aspuru-Guzik* groups, *Nelson* performed the first experimental QPT in a "warm" and complex system, using double-walled J-aggregate nanotubes.

The QPT technique will be the basis of our ongoing research efforts in quantum transport. Notably, we plan to extend our spectroscopic capabilities and build the first entangled 2d Fourier Transform Spectroscopy tool, which should enable us to significantly reduce the optical power required to probe exciton interactions in new samples, such as the covalent organic frameworks made by *Dinca* and characterized by *Black*, or the valley coherences in WSe_2 studied by *Levitov* and *Jarillo-Herrero*. We will also exploit our control over exciton coherence to improve exciton transport without the manufacturing challenge and expense of removing disorder using "topological excitonics".

Center for Excitonics (CE)	
Massachusetts Institute of Technology	Marc Baldo, Moungi Bawendi, Vladimir Bulovic, Mircea Dinca, Dirk Englund, Silvija Gradecak, Pablo Jarillo-Herrero, Jing Kong, Leonid Levitov, Keith Nelson, Will Tisdale, Troy Van Voorhis.
MIT Lincoln Laboratory	Eric Dauler
Harvard University	Alan Aspuru-Guzik
Brookhaven National Laboratory	Charles Black, Eric Stach

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Solid-State Solar-Thermal Energy Conversion Center (S^3TEC)
EFRC Director: Gang Chen
Lead Institution: Massachusetts Institute of Technology
Start Date: August 2009

Mission Statement*: To advance fundamental science and to develop materials for harnessing heat from the Sun and terrestrial sources and converting this heat into electricity via solid-state thermoelectric, thermophotovoltaic, and thermogalvanic technologies.*

The DOE EFRC center entitled "Solid-State Solar-Thermal Energy Conversion Center (S^3TEC Center)" is formed by the Massachusetts Institute of Technology, in partnership with Boston College, the California Institute of Technology, the University of Houston, Brookhaven National Laboratory, and Oak Ridge National Laboratory. The S^3TEC Center was founded in 2009 with the vision to create an intellectual base for advancing the fundamental science of energy-carrier coupling and transport for thermal-to-electrical energy conversion, originally based on thermoelectric (TE) and thermophotovoltaic (TPV) technologies.

These heat-to-electricity energy conversion technologies represent a drastic departure from conventional thermal-mechanical energy conversion engines. They are solid-state and do not involve intermediate mechanical energy conversion. They are attractive for their compactness, scalability, and environmental friendliness, but currently are not efficient enough to compete against conventional energy conversion technologies.

For the last five years, the fundamental knowledge gained at the S^3TEC Center has been successfully used to design new materials and to demonstrate principle-of-principle prototypes to achieve leaps in efficiency of these technologies, with an emphasis on approaches that have the potential to be low-cost. The S^3TEC Center's focus on fundamental studies of the transport of electrons, phonons, and photons, and the interactions among these carriers has led to improvements in material performance and promising technology demonstrations, including factors of 7-8 and 3 higher efficiencies of solar TE and solar TPV generators compared to the state-of-the-art, respectively.

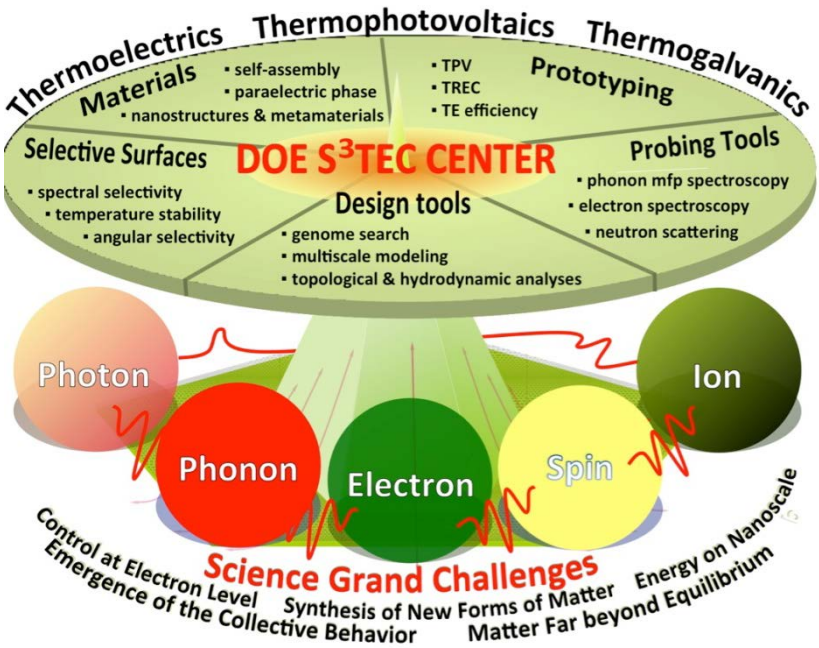


Figure 1. Vision of S^3TEC Center. The Center focuses on electron and spin, ion, phonon, and photon transport and interactions underpinning direct heat-to-electricity energy conversion based on thermoelectric, thermogalvanic, and thermophotovoltaic technologies. Addressing grand challenges yields new materials and methods for simulation and characterization.

The renewed S^3TEC Center will build on the progress made and expand its research horizons: energy sources to be harnessed will include not only solar but also terrestrial heat; in addition to TE and TPV, thermogalvanics (TG) will join the list of pursued technologies through our proposed study on thermally regenerative electrochemical

cycles (TREC); the fundamental energy carriers studied will include not only electrons, phonons, and photons, but also ions and spin; the emphasis on computational material design and mesoscale simulation to guide material synthesis will be increased. Figure 1 illustrates the vision behind the S^3TEC center and the scope of research it encompasses.

TE, TREC, and TPV technologies have high potential for a disruptive transformation in our efforts to harness an almost endless energy supply from the sun and terrestrial heat sources. However, successful development of these technologies can be achieved only through fundamental breakthroughs in materials design and understanding of transport processes. While we know to a certain degree how to control photons, we know much less about how to control electrons and ions in this context, and very little about how to control phonons. Many fundamental questions and challenges lie ahead, and to address them, the S^3TEC Center will strive to take our fundamental understanding to the logical next step: to designing materials and processes that can be scaled up, so that research can be applied to meet the energy challenges of our nation and the world.

The Center aims to address the DOE's grand challenges, connecting nanoscale materials to gigawatts electricity, by advancing the basic science underpinning direct energy conversion technologies, through discovering materials that meet the needs of applications in terms of performance, cost, and reliability. By studying TE, TG, and TPV technologies in one Center, great synergy is generated in basic science, technology, and applications. The Center will advance and employ photon, neutron, and electron beams and first-principles simulations tools to enable the probing and understanding of the transport processes of the carriers and the prediction of their properties relevant for energy conversion.

This understanding will aid in the design and synthesis of materials to validate the theory and construct proof-of-principle prototypes demonstrating the potential to achieve high heat-to-electricity conversion efficiency. The S^3TEC Center will build on its established management structure which continues to encourage and strengthen synergistic collaborations, nurture future leaders in energy technology, and engage the public through outreach activities.

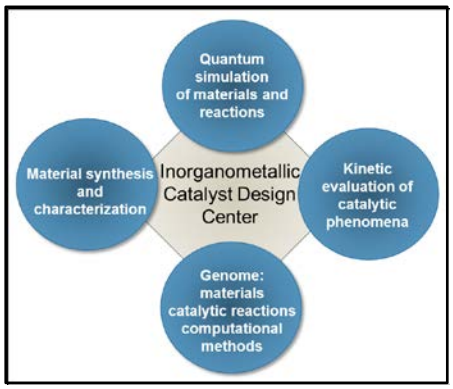
Solid State Solar Thermal Energy Conversion Center (S^3TEC)	
Massachusetts Institute of Technology	Gang Chen (Director), Evelyn N. Wang (Associate Director), Mildred Dresselhaus, Marin Soljacic, Keith Nelson, Yang Shao-Horn, Caroline Ross, Nicolas Hadjiconstantinou, Alexie Kolpak
University of Houston	Zhifeng F. Ren
Oak Ridge National Laboratory	David J. Singh, Olivier Delaire
California Institute of Technology	Jeffrey Snyder
Boston College	David Broido
Brookhaven National Laboratory	Yimei M. Zhu

Contact: Prof. Evelyn N. Wang, Associate Director, enwang@mit.edu
617-324-3311, http://s3tec.mit.edu/

Inorganometallic Catalyst Design Center (ICDC)
EFRC Director: Laura Gagliardi
Lead Institution: University of Minnesota
Start Date: August 2014

Mission Statement*: To computationally guide the discovery of a new class of energy-science-relevant catalytic materials and understand the underlying structure-function relationships that will guide further catalyst discovery.*

The ICDC EFRC will design a new class of catalysts consisting of precisely synthesized inorganometallic clusters ("nodes") with <200 atom, (usually <40 atoms of each element), supported in uniform arrays by organic linkers based on metal-organic frameworks (MOFs) or surface-supported porphyrins. Each node will be identical, and designed with atomic precision to incorporate up to three functional materials simultaneously: bimetallics, mixed oxides (or sulfides) and even support oxides. Size effects of the oxide (or sulfide) components of these cluster materials will be studied for the first time. We will computationally predict such cluster catalysts for conversions of natural gas into methanol and oligomers, investigating their reactivity with new quantum chemical and classical simulation techniques. We will validate and employ new methods for the computational screening and ranking of thousands of hypothetical catalysts. In parallel with the computational design, new synthetic approaches will be used to assemble, sometimes atom-by-atom, high-density, uniform three-dimensional arrays of isolated candidate clusters rapidly, and in amounts sufficient for characterization and performance testing. These new materials will be structurally characterized and evaluated for adsorption properties and catalytic activity, selectivity, and stability. We envision the development of fundamentally new and superior dispersed sub-nanometer catalysts for energy-intensive, energy-relevant chemical reactions—especially reactions that must move to the forefront of energy technology and environmental protection because of their direct connection to the accelerating natural gas boom.

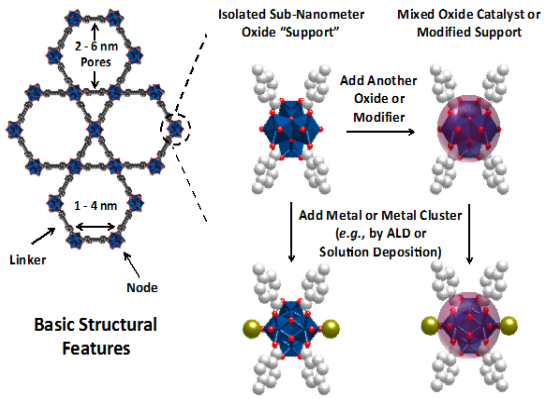


Main elements of ICDC.

The core of the ICDC will be advanced modeling for in silico design of new catalysts of potentially extraordinary activity and selectivity. Our central premise is that we can successfully design catalytic sites by focusing on subnanometer and few-nanometer sized clusters of atoms that constitute both the catalytically active species and support-like components within the same cluster. This design approach will be accelerated by structural, energetic, and mechanistic modeling coupled with selective, high-throughput computation, with the aim of identifying better and better candidates. No less important will be a deepening of fundamental understanding. The ability to test hypotheses in silico over an enormous and arbitrarily modifiable database of hypothetical catalysts is expected to lead to compelling, transferrable catalyst design rules and potentially lead to the discovery or design of new mechanisms and desirable new catalytic chemistry. Computational design, screening, and synthesis will be complemented by high-throughput experimental screening enabled by the expertise and instrumentation developed by our team members at Dow. Dow will also provide essential expertise regarding industrial needs in terms of both catalyst performance and requisite basic understanding.

The mission of the ICDC encompasses developing supported cluster catalysts with some or all of the following characteristics:

- Uniform mesoscale structures that can be synthesized reproducibly, which are ideal for both full experimental structural characterization and computational modeling.
- Clusters that are well defined and uniform in structure and composition, but isolated from each other on supports to minimize cluster-cluster interactions and agglomeration.
- Catalyst structures that are amenable to high-throughput experimental screening, very reliable quantum mechanical predictions on prototype exemplars.
- Catalytic properties that can be modulated by cluster size, with separate control over the size of both the catalytic cluster and the support cluster to which it is attached, and compositions that are not subject to the restrictions of bulk-material-phase thermodynamics.
- Tight single-atom control and access to pre-defined cluster compositions, extending even to nominally naked clusters without residual capping ligands.
- Compatibility with a wide range of property-modifying support compositions.
- Robustness, including resistance to aggregation/agglomeration.



Schematic of the new class of materials proposed.

The samples will span the range from nano- to mesoscale structures, as the catalytic species will be clusters, some consisting of only a few atoms, and the supports will be mesoporous materials, including metal-organic frameworks (MOFs). With regard to the computational predictions, we will develop improved theoretical methods for accurate modeling of the reactivities and synthetic likelihoods, as well as the catalytic properties of the target materials, again building upon recent pioneering developments by team members. Our ultimate goal is to design and produce catalysts for reactions that will yield significant energy savings and environmental protection relative to existing alternatives, but we recognize that in the time span of the proposed research these goals are highly ambitious, and a more immediate outcome of the research (and one that may be even more important in the long term) will be what we learn about structure-function relationships for new catalysts in this important mesoscale size range.

Inorganometallic Catalyst Design Center (ICDC)	
University of Minnesota	Laura Gagliardi (Director), Donald Truhlar (Theme Leader), Christopher Cramer, Connie Lu, R. Lee Penn, Andreas Stein
Northwestern University	Joseph Hupp (Deputy Director), Omar Farha, Randy Snurr
University of Washington	Charles Campbell (Theme Leader)
University of California-Davis	Bruce Gates (Theme Leader)
Pacific Northwest National Laboratory	Johannes Lercher (Theme Leader)
Argonne National Laboratory	Karena Chapman, Alex Martinson
Clemson University	Rachel Getman
Dow Chemical Company	Edward Calverly

Contact: Laura Galiardi, Director, gagliard@umn.edu
612-625-8299, http://www.chem.umn.edu/icdc/

Biological Electron Transfer and Catalysis (*BETCy*)
EFRC Director: John Peters
Lead Institution: Montana State University
Start Date: August 2014

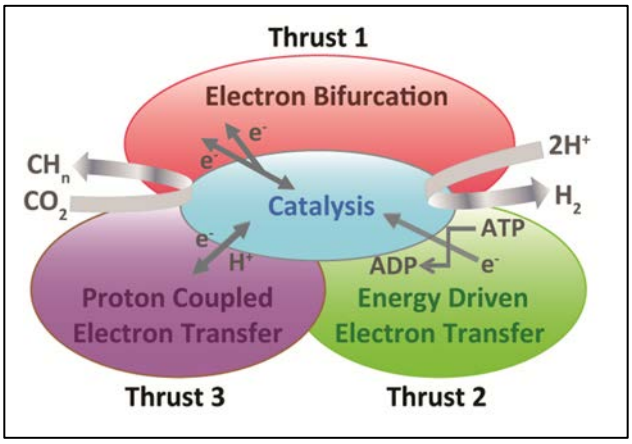
Mission Statement*: To investigate the mechanisms and structural basis controlling electron bifurcation, electron-ion coupling, and redox catalysis in model enzymes; this detailed understanding of how biological systems control and exploit electron flow will be used to develop modular biochemical conversions for the production of hydrocarbon and hydrogen biofuels.*

This project will conduct an in-depth study of metalloenzyme structure-function relationships that enable retuning of catalytic sites, electron-transfer thermodynamics, and the evolution of tailor-designed chemical reactions involving two model enzymes: hydrogenase and nitrogenase. We propose studies in three Thrust areas to realize these goals: **Thrust 1.** Energy conservation and electron flow; **Thrust 2.** ATP-coupled electron transfer; and **Thrust 3.** Atomic level determinants of enzymatic redox properties and their relationship to catalytic bias. We will develop a collective knowledge in metalloenzymes by applying physical science and computational tools to characterize biochemical reactions catalyzed specifically by multi-subunit enzymes harboring iron-sulfur clusters and flavin cofactors, as models for redox reactions throughout biology. Understanding these mechanisms is central to overcoming the key thermodynamic barriers currently limiting production of reduced products and biofuels.

Biological systems are not typically under selective pressure to optimally and maximally produce an overabundance of highly reduced products such as advanced biofuels and chemicals as well as their precursors. Generating organisms with these energy relevant characteristics requires biotechnological manipulation guided by a strong fundamental understanding of key electron flow mechanisms. One significant limitation for the production of highly reduced bioproducts is that a large proportion of the metabolic electron flow is directed towards reduction of pyridine nucleotides (i.e., to NAD(P)H). Thermodynamically the reduced pyridine nucleotides are not sufficient in energy for the production of the desired highly reduced products. A newly discovered biochemical mechanism termed “electron bifurcation” potentially overcomes this thermodynamic challenge by combining electrons from the reduced pyridine nucleotides with other lower potential electrons from reduced Ferredoxins (Fd). This increases the reducing potential of electrons from reduced pyridine nucleotides such that a larger flux is directed toward the production of desired reduced products. Electron bifurcation is still poorly understood and a more detailed understanding of this phenomenon could lead to “game changing” advances in engineering model organisms that can optimally direct electron flow to reduced products. Furthermore, such a knowledge base may provide a blueprint for bio-inspired, multi-electron catalytic processes that can utilize electrons of varying reduction potentials.

The focus of the Biological Electron Transfer and Catalysis (*BETCy*) EFRC is to provide a fundamental understanding of mechanisms that allow biology to overcome key thermodynamic barriers that limit the production of reduced products. The three interrelated research Thrusts of the *BETCy* EFRC (depicted in the figure below) emphasize mechanisms of electron bifurcation and energy conservation (**Thrust 1**), as well as ATP- (**Thrust 2**) and proton-coupled electron transfer (**Thrust 3**) mechanisms. The proposed research of the *BETCy* EFRC builds on recent seminal discoveries and provides the basis for attacking key knowledge gaps and expanding the knowledge base that is essential for realizing the true potential for bioenergy as a prominent component of the global energy portfolio. We have assembled a strong team of investigators with complementary research interests and technical skills to accomplish the proposed

interdisciplinary tasks. The work will have profound scientific impact generating a blueprint for efficient control of electron flow into energy products and chemicals. The proposed work is directly in line with three of the five Basic Energy Sciences Advisory Committee (BESAC) Grand Challenges including: 1. Control of material processes at the level of electrons, 2. Design and perfect atom and energy efficient synthesis of revolutionary new forms of matter with tailored properties 3. Characterize and control matter away, far away, from equilibrium. In addition, the proposed *BETCy* EFRC goals embrace the Basic Research Needs (BRNs) outlined in the BES Workshop Report on "Catalysis for Energy", and makes strong connections with the "Hydrogen Economy" and "Solar Energy Utilization" BRNs.



The research plan of the *BETCy* EFRC has been designed to provide the basis to exploit unique biochemical mechanisms that have yet to be explored substantively in the context of bioenergy but have the potential for game-changing advancement. The successful outcome of this research will be a collection of fundamental principles that serve as a blueprint to enable the tailored re-design of biological systems and enzymes to control matter and energy at the level of electrons and molecules to provide the foundations to create new energy technologies.

Center for Biological Electron Transfer and Catalysis (BETCy)	
Montana State University	John W. Peters (Director), Brian Bothner, Eric Boyd, Ross Carlson
University of Georgia	Michael W.W. Adams (Associate Director of Research)
National Renewable Energy Laboratory	PinChing Maness (Associate Director of Project Management), Paul King
University of Washington	Caroline Harwood
Arizona State University	Anne Jones
Utah State University	Lance Seefeldt
University of Kentucky	Anne-Frances Miller

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Center for Next Generation of Materials by Design (CNGMD): Incorporating Metastability
EFRC Director: William Tumas
Lead Institution: National Renewable Energy Laboratory
Start Date: August 2014

Mission Statement*: To dramatically transform the discovery of functional energy materials through multiple-property search, incorporation of metastable materials into predictive design, and the development of theory to guide materials synthesis.*

The Center for Next Generation of Materials by Design (CNGMD) EFRC aims to discover and synthesize novel functional materials including non-equilibrium structures in a predictive manner. The specific goals of the CNGMD EFRC are to:

1. Design and discover new energy-relevant materials with targeted functionalities by integrating theory, high-throughput computation, synthesis, and characterization.
2. Develop foundational theoretical, synthesis, and characterization tools to enable the next generation of Materials by Design.
3. Incorporate functional metastable materials into Materials by Design and establish ranges for materials metastability as a function of the chemistry and structure arising from defects, polymorphs, and solid solutions (alloys).
4. Develop a systematic theory-driven approach to guide the synthesis of new materials—including metastable systems—by coupling theory and state-of-the-art *in-situ* characterization to probe materials growth pathways.
5. Promote and disseminate the Next Generation of Materials by Design to the broader materials science community.

To realize the ***Next Generation of Materials by Design***, we will couple first-principles theory with state-of-the-art synthesis and characterization to understand and predict structure, properties, and phenomena at the molecular, nano, and meso scales. This EFRC is specifically designed to address several critical scientific gaps that must be overcome before computational materials design becomes a robust tool that delivers new functional materials — namely 1) ***multiple-property design***, 2) ***accuracy and relevance***, 3) ***metastability***, and 4) ***synthesizability***. This approach will expand and accelerate the discovery of experimentally accessible materials for energy applications.

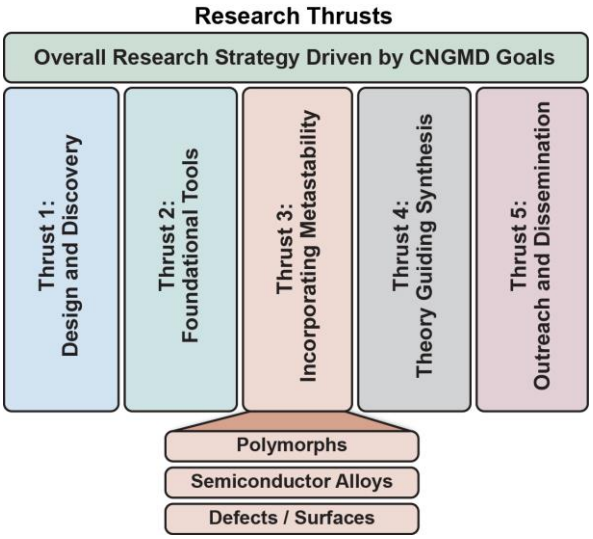

Fig. 1: Five Research Thrusts for our EFRC, each co-led by a theorist and an experimentalist.

Our initial focus will be on semiconductor materials for solar energy conversion, solid-state lighting, and related technologies—all areas that are in need of transformative materials. Specific studies will concentrate on new materials for photo-active absorbers, transparent conductors, contacts and electrodes. The newly developed methodologies will

also impact the broader area of materials discovery including other areas of energy conversion, storage or utilization. Our research will be implemented through five thrusts (Fig. 1), each with synergistic theory and experimental components.

Thrust 1–Design and Discovery integrates high-throughput computing and experiment into a rapid design methodology that recognizes the need for simultaneous searching over multiple properties. We will develop an *integrated theory and experiment approach for accelerating materials design* that we will broaden to include a predictive capability including metastability.

Thrust 2–Foundational Tools Thrust will improve or develop the critical theoretical and experimental capabilities including advanced theoretical methods for rapidly predicting atomic and electronic structure, novel *in-situ* characterization, and high-throughput synthesis and characterization techniques.

Thrust 3–Incorporating Metastability focuses on developing a fundamental understanding of metastability required to rationally and successfully incorporate non-equilibrium materials into Materials by Design. We will investigate three classes of metastability that are specifically relevant to inorganic semiconductors for optoelectronic applications: ***polymorphism***; ***defects, disorder, and interfaces***; and ***semiconductor alloys***.

Thrust 4–Theory Guiding Synthesis aims to transform computational materials design by predicting the experimental conditions required to synthesize the new functional materials. Predicting **how** to grow a desired material is the next most important aspect after predicting **what** material is worth growing. To begin, we will focus on homogeneous solution precipitation, electro-deposition, and vapor-phase deposition. We seek to understand the thermodynamics of intermediate states (e.g., nucleation) as well as transient and local conditions (e.g., high chemical potentials of species established by decomposition of reactive precursors) to explain which polymorphs, defects, and alloys form under particular synthesis conditions. The stability and synthesis space for new, not previously synthesized materials will be investigated using advanced *in-situ* and atomistic characterization methods. This will elucidate the mechanisms of growth and experimental measurements and will benchmark newly developed theoretical models.

Thrust 5–Outreach and Dissemination aims to build and assist the Materials by Design community and to reach out to broader materials science groups by making our methodology and data accessible.

Center for Next Generation of Materials by Design (CNGMD)	
National Renewable Energy Laboratory	William Tumas (Director), David Ginley (Chief Experimentalist), John Perkins (Program Integrator), Stephan Lany, Andriy Zakutayev
Massachusetts Institute of Technology	Gerbrand Ceder (Chief Theorist), Alexie Kolpak, Tonio Buonassisi
Lawrence Berkeley National Laboratory	Kristin Perrson
SLAC National Accelerator Laboratory	Michael Toney
Harvard University	Daniel Nocera, Roy Gordon
Oregon State University	Janet Tate, John Wager
Colorado School of Mines	Vladan Stevanovic, Brian Gorman

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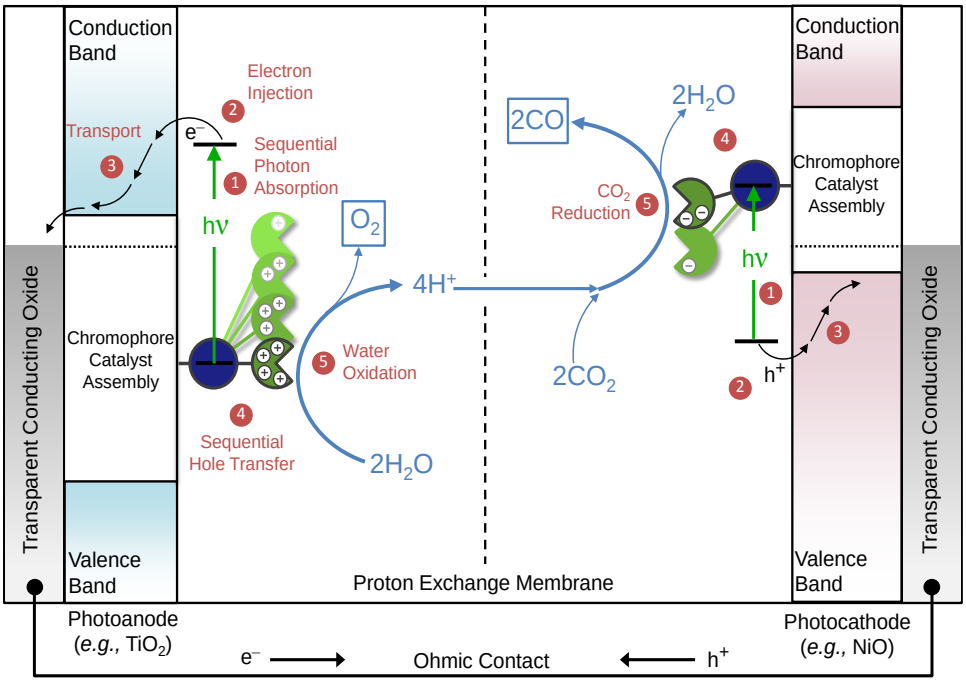
Center for Solar Fuels (UNC)
EFRC Director: Thomas J. Meyer
Lead Institution: University of North Carolina at Chapel Hill
Start Date: August 2009

Mission Statement: *To conduct research on dye sensitized photoelectrosynthesis cells (DSPECs) for water splitting and tandem cells for the reduction of carbon dioxide to carbon-based solar fuels.*

The UNC EFRC Center for Solar Fuels is conducting research on solar energy conversion and storage by using the sun to drive solar fuel reactions. Center research extends from basic research on fundamental processes and new materials to integrating components into surface-stabilized assemblies, and assemblies into device prototypes. In its research the Center utilizes a broad, multidisciplinary approach in a highly collaborative setting. It draws on expertise in chemistry, physics, and materials science with a focus on the concept of the Dye Sensitized Photoelectrosynthesis Cell (DSPEC) for solar fuels production.



TARGET METRICS

- Converting solar photons into redox equivalents in the attached chromophore-catalyst assembly with >90% efficiency (steps **1**, **2**).
- Building up multiple redox equivalents at catalyst sites for water oxidation or CO_2 reduction with efficiencies >95% on the timescale of the light source, seconds for ambient sunlight (steps **3** and **4**).
- Achieving catalytic rates of $>10^2$ s^{-1} (step **5**).
- Achieving overall device efficiencies of 15% with 10^7 cycles/year and a 95% retention in photoreactivity.

Dye-Sensitized Photoelectrosynthesis Cell (DSPEC) for CO_2 Reduction to CO

THE DYE-SENSITIZED PHOTOELECTROSYNTHESIS CELL (DSPEC) FOR SOLAR FUELS PRODUCTION

Research on high efficiency DSPEC devices in the Center for Solar Fuels builds on a modular approach. Catalysts for water oxidation or CO_2 reduction, light-harvesting chromophores and chromophore arrays, chromophore-catalyst assemblies, mesoporous nanoparticle semiconductor oxide and transparent conducting oxide films, and core/shell structures are all investigated separately and integrated in thin film structures for possible DSPEC applications. Atomic layer deposition (ALD) is used to create core/shell structures and to stabilize surface-bound assemblies. Interfacial assembly structures are characterized dynamically by transient spectroscopic measurements on timescales from femtoseconds to seconds. Interfacial chromophore-catalyst assemblies surface-stabilized on core/shell oxide structures provide the basis for DSPEC photoanodes and photocathodes. In a breakthrough, research in the Center has led to working DSPECs for sustained water splitting with a small applied bias. Extensions, with modifications in core/shell structures using ALD or surface polymer stabilization, hold promise for enhanced efficiencies and long term stabilities under operating device conditions.

Future progress will build on the results of catalyst developments in the initial grant period with a focus on catalysis by earth abundant elements. The targets are catalysts that are stable through 100,000s of turnovers at rates that exceed 10 s^{-1}. Catalysts will be integrated with organic and metal complex chromophores for efficient solar harvesting while meeting the energetic demands of the catalyzed reactions. Molecular assemblies that link multiple chromophores to a single catalyst will be exploited based on polymeric scaffolds and peptide assemblies. The successful core/shell photoanode architecture will be extended to new materials and new core/shell configurations to maximize solar efficiencies, and to *p*-type oxides for photocathode and tandem cell applications. ALD, electropolymerization, and polymer overlayer strategies will be used to stabilize surface structures and control interfacial properties and dynamics. Finally, existing steady-state and time-resolved experimental protocols will be used to quantify energy conversion efficiencies and investigate long term stabilities under DSPEC operating conditions.

Research in the Center for Solar Fuels is multidisciplinary and highly integrated. The challenges posed by the modular approach are met by an integrated, team-based structure with the current teams listed below. Future research will build on this structure, existing UNC EFRC infrastructure, and research progress to date to develop stable, efficient DSPEC prototypes for water splitting with extensions to tandem prototypes for simultaneous water oxidation and CO_2 reduction. Based on past success, the Center is poised to have, what may be, a transformative impact on solar energy conversion and storage.

RESEARCH	TEAM	TEAM RESEARCH FOCUS
Catalysis	**Water Oxidation**	Development and mechanistic studies of interfacial catalysts for water oxidation; integration of catalysts into assembly structures, and device prototypes.
	CO_2 Reduction	Development and mechanistic studies of interfacial catalysts for CO_2 reduction; integration of catalysts into assembly structures, and device prototypes.
Assemblies	**Molecular Assemblies**	Synthesis & surface binding of chromophore-catalyst assemblies at metal oxide interfaces.
	Polymers	Synthesis and characterization of molecular assemblies based on polymer scaffolds for multi-chromophore and chromophore-catalyst applications.
The Interface	**Interface Dynamics**	Dynamics of light-driven interfacial electron transfer in chromophores, assemblies, and chromophore-catalyst assemblies on metal oxide semiconductor surfaces.
	Interface Structure and Stabilization	Characterization of surface structure and binding of molecular chromophores, catalysts and assemblies surface-bound to metal-oxide semiconductors. Stabilization of surface structures by ALD, electropolymerization, and polymer film overlayers.
Device Prototypes	**Semiconductors and Devices**	Semiconductor, transparent conducting oxide, and core/shell structures for DSPEC applications. Performance of integrated photoelectrodes in DSPEC device prototypes.

Center for Solar Fuels (UNC)	
University of North Carolina at Chapel Hill	T.J. Meyer (Director), J.M. Papanikolas (Co-Deputy Director), G.J. Meyer (Co-Deputy Director), M.S. Brookhart, J.F. Cahoon, J.L. Dempsey, G.L. Glish, Y. Kanai, R. Lopez, A.J.M. Miller, A.M. Moran, R.W. Murray, C.K. Schauer, J.L. Templeton, M.L. Waters, L. Alibabaei, M.K. Brennaman
Georgia Institute of Technology	J.R. Reynolds
University of Florida	K.S. Schanze
University of Colorado, Boulder/NREL	A.J. Nozik
Research Triangle Institute	P.G. Hoertz

Contact: Thomas J. Meyer, Director, Center for Solar Fuels, tjmeyer@unc.edu
919-843-8313, www.efrc.unc.edu

The Center for Bio-inspired Energy Science (CBES)
EFRC Director: Samuel I. Stupp
Lead Institution: Northwestern University
Start Date: August 2014

Mission Statement*: To discover and develop bio-inspired systems that reveal new connections between energy and matter.*

The goal of the Center for Bio-inspired Energy Science (CBES) is to develop artificial materials and systems that take inspiration from biology to optimize the way we use energy and interconvert between different energy forms, for example converting chemical energy into mechanical energy the way muscles do in living organisms. Our team members have innovated in the areas of self-assembly, the interface of biology and materials science, as well as in theory and simulation. The proposed research addresses the following two DOE grand challenges: "how do we characterize and control matter away-especially very far away-from equilibrium?" and "how can we master energy and information on the nanoscale to create new technologies with capabilities rivaling those of living things?" Work proposed also addresses DOE's basic research need for "new science for a secure and sustainable energy future." CBES is scientifically organized into three main thrusts with many inter-thrust connections, which will increase and strengthen throughout the existence of the Center. We expect strong inter-connectivity among all three groups, yielding outstanding scientific synergy for discovery and ideas.

In **Thrust 1** we focus on artificial materials that show functions that emulate the fundamental properties of cells, extracellular matrices, and tissues in the context of energy use and transduction. Our team has identified three primary bio-inspired research areas: artificial muscles, artificial organelles, and stimulus-driven adaptive materials. The work on **artificial muscles** is focused on systems that can interconvert between chemical and mechanical energy forms (as muscles do). The challenge of discovering the right strategies to transduce stored chemical energy into mechanical motion in materials will require not only synthetic innovation in soft matter but also the use of computational models to find structures in which small energy inputs result in large deformations and forces. In this work, we will take advantage of the great computational and theoretical capabilities of the team and also of the enormous recent progress made in DNA nanotechnology to help us discover muscle-like structures in polymer-nucleic acid hybrid materials. Inspired by biological organelles that utilize feedback mechanisms to mediate chemical reactions, we will develop **artificial organelles** that have the capacity to mediate efficiently synthetic reactions as cells do and in ways that chemical laboratories and chemical factories cannot at present. For this purpose, we have planned experiments with precisely positioned micro-particles mediating related chemical events with the goal to discover emergent principles for self-regulating reactions. Another interesting characteristic of biological matter is the ability to dynamically change shape in response to the environment. This characteristic is possibly most prominent in the changes undergone by cells during differentiation, proliferation, and migration, using energy inputs to achieve these functions. In the context of bio-inspiration we have defined this problem as the challenge of developing **adaptive materials** that respond to stimuli such as light and mechanical forces to mutate their structures and properties.

In **Thrust 2** we explore the bottom-up conversion of energy to motion in artificial nanometer to micrometer scale colloids that behave collectively far from equilibrium. Miniaturizing machines down to the nanoscale raises both fundamental questions and novel opportunities to perform tasks that mimic or improve upon biological machines. Unlike most synthetic machines, which rely on centralized engines, biological organisms convert energy into motion in a highly *distributed* fashion: macroscopic

forces emerge from the coordinated actions of many chemically powered, molecular scale actuators (motor proteins). Distributed energy conversion in biology offers several advantages that motivate the pursuit of much simplified *non-biological* colloidal machines, in which energy is harnessed at colloidal scales to produce emergent behaviors at the mesoscale and perform useful functions at the macroscale. The proposed work aims to significantly extend these capabilities to create **colloidal motors** capable of increasingly complex locomotive forms, to control motor activity remotely through external stimuli, to improve the efficiency of colloidal motors by many orders of magnitude, and to engineer switchable interactions among colloidal motors to guide their collective behaviors. By combining computer simulation, continuum modeling, and a suite of experimental systems spanning scales from nano- to macroscopic, the team aims to discover how the distributed activity of **ensembles of colloidal motors** produce emergent collective behaviors. Once again, cellular behavior inspires our design. Cells undergo dramatic changes in size and shape; they crawl or swim through diverse environments; they capture and transport external cargo; they grow, proliferate, and differentiate; they self-organize to form tissues and organs. Each of these remarkable capabilities relies on the coordinated action of many individual actuators distributed throughout the cell. With this inspiration, the proposed work aims to create colloidal machines that mimic – in rudimentary form – the mechanical functions of living cells. These non-biological, **artificial cells** will be composed of many colloidal motors within a flexible membrane through which chemical fuel is delivered. Through the collective motions of the confined motors, we envision microscale artificial cells capable of *controllable deformations*, *switchable mechanical properties*, and *autonomous locomotion*. Hierarchical assemblies of these "cells" will produce macroscopic, non-equilibrium materials that harness distributed energy conversion and actuation to perform useful functions.

Another critical axis of energy management in biology is connected with modes of charge transport that are not known in artificial matter, such as long-range electron transfer in photosynthesis through media of low conductivity or the selective ionic fluxes across cell membranes that can convert chemical to electrical energy. Therefore in **Thrust 3** we focus sharply on bio-inspired energy and charge transport modes. We are particularly interested in solar energy conversion systems comprising low-dielectric active materials, in which the major obstacle to efficient energy conversion is a low yield of charge collection due to random, rather than directional, motion of electrons. We will address this challenge by exploring, computationally and experimentally, the non-equilibrium phenomenon of **quantum ratcheting**. We are also exploring how to control selectivity and directionality of non-equilibrium ion fluxes by theoretically and experimentally designing nanoscale **bio-inspired ion pumps**.

Center for Bio-inspired Energy Science (CBES)	
Northwestern University	Samuel Stupp (Director), Bartosz Grzybowski, Chad Mirkin, Monica Olvera de la Cruz, Mark Ratner, George Schatz, Igal Szleifer, Emily Weiss
University of Pittsburgh	Anna Balazs
Pennsylvania State University	Kyle Bishop
New York University	Paul Chaikin
University of Michigan–Ann Arbor	Sharon Glotzer
Harvard University	George Whitesides

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Argonne-Northwestern Solar Energy Research (ANSER) Center
EFRC Director: Michael R. Wasielewski
Lead Institution: Northwestern University
Start Date: August 2009

Mission Statement*: To revolutionize our understanding of molecules, materials, and methods necessary to create dramatically more efficient technologies for solar fuels and electricity production.*

The ANSER Center is fulfilling this mission by developing a fundamental understanding of solar energy conversion dynamics, by designing and synthesizing new nanoscale architectures with extraordinary functionality, and by linking basic solar energy conversion phenomena across time and space to create emergent energy conversion systems operating with exceptional performance. The ANSER Center addresses a common set of fundamental questions that must be answered to successfully utilize sunlight as a renewable energy source for fuels and electricity. By attacking the common questions intrinsic to solar fuels and electricity in the same research center, the ANSER Center provides opportunities for cross-cutting solutions not possible by addressing only fuels or electricity alone:

- How can multi-scale predictive theory and computational modeling lead to the design and discovery of novel organic, inorganic, and hybrid systems?
- How do molecular and materials structure and order determine the efficiency of light capture, charge separation, and long-range charge transport?
- What are the fundamental multi-scale temporal and spatial requirements for efficient charge transport across interfaces to deliver multiple redox equivalents to catalysts and electrodes?
- How can molecular and materials properties be tailored to exploit hierarchical assembly for solar fuels and electricity systems scalable from the nanoscale to the mesoscale?

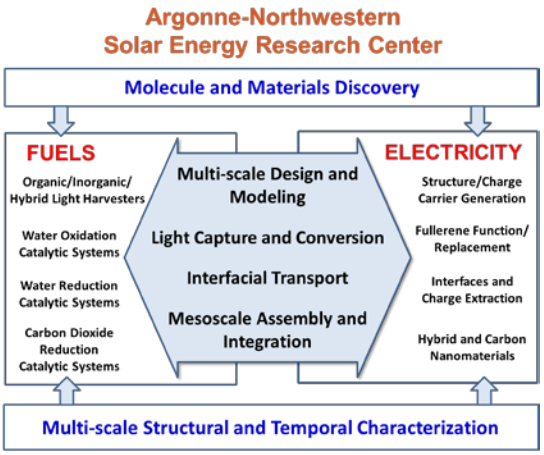

Fig. 1. The ANSER Center integrated research program is built on the strengths of molecule and materials discovery and multi-scale structural and temporal characterization.

ANSER Center strengths include molecule and materials discovery and high-resolution spatial and temporal analytical techniques and tools. Specialized X-ray facilities, such as the Advanced Photon Source at Argonne National Laboratory, provide atomic-scale structural information using state-of-the-art *in situ* X-ray absorption and scattering techniques. Ultrafast time-resolved absorption, emission, and vibrational spectroscopy along with time-resolved electron paramagnetic resonance (EPR) spectroscopy and appropriate computational simulation and modeling at ANSER Center laboratories provide detailed information on mechanisms and dynamics.

Subtask 1: Molecules, Materials, and Systems for Solar Fuels. Our greatest challenge is efficient fuel production at acceptable rates and driving forces. The ANSER Center is employing a hierarchical approach to understanding catalyst and photocatalyst function; thus, requiring a level of integration that cannot be achieved by any single research group. ANSER Center catalyst discovery is strongly hypothesis-driven, nicely complementing but not duplicating the approach of our collaborator, the DOE Joint Center for Artificial Photosynthesis (JCAP) Energy Innovation Hub. The ANSER Center emphasizes catalysts ranging from molecules, to clusters, to nanoparticles, to bulk materials that: a) are derived

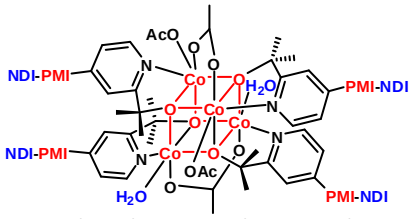

Fig. 2. Co-based water oxidation catalyst with appended light-harvesting, charge-generating perylenemonoimide-naphthalendiimide (PMI-NDI) donor-acceptor ligands.

from plentiful elements, b) have promising compositions that are thermodynamically inaccessible as bulk (macroscopic) materials, and c) may enable access to new mechanistic pathways, thereby moving beyond perceived fundamental or practical limits on catalyst performance, including catalyst kinetics and overpotentials. Solar fuels research is exploring and exploiting new ideas about: a) organic/inorganic/hybrid light harvesters, b) water oxidation catalysis (e.g. Figure 2), c) water reduction catalysis, and d) carbon dioxide reduction catalysis.

Subtask 2: Molecules, Materials, and Systems for Solar Electricity. Photovoltaic cells fabricated from relatively simple, non-toxic, earth-abundant, mechanically flexible, and low-cost materials offer the prospect of efficient large-scale solar electricity production. Efficiencies have advanced dramatically in the past five years, driven by an ever-increasing, but by no means complete, understanding of the relevant chemistry, materials science, physics, and performance limits. A closely integrated, highly productive interplay of synthesis, characterization at multiple time and length scales, and theoretical analysis and prediction, led to a number of ANSER Center "firsts" in materials design, mechanistic understanding, and performance metrics. Following the same collaborative strategies, ANSER Center solar electricity research continues to explore and exploit new ideas about: a) perovskite-based hybrid solar cells (e.g. Figure 3), b) active layer polymer/small molecule structure-exciton dynamics, c) fullerene acceptor uniqueness and designed replacements, d) new interfacial materials and phenomena, and e) unconventional carbon nanomaterial and metal-organic active layers. ANSER Center research is testing ideas driven by theory to understand at a fundamental level how photovoltaic cell performance is affected by nanoscale/mesoscale architectural-electronic structure relationships in soft-matter and in hybrid soft-matter/hard-matter solar cells, photon capture, exciton creation and decay, exciton dissociation/quenching, and charge transport, electrode microstructure, doping, and surface chemistry. Understanding these phenomena feeds back directly to developing light capture and charge delivery strategies to power solar fuels catalysts as well.

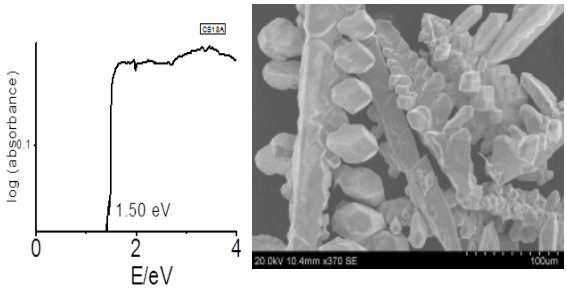

Fig. 3. Optical absorption spectrum (left) and crystals (right) of (formamidinium)SnI_3, a lead-free organic-inorganic hybrid perovskite.

Argonne-Northwestern Solar Energy Research (ANSER) Center	
Northwestern University	Michael Bedzyk, Robert Chang, Mark Hersam, Joseph Hupp, Mercouri Kanatzidis, Tobin Marks, Mark Ratner, George Schatz, Samuel Stupp, Michael Wasielewski (Director), Emily Weiss
Argonne National Laboratory	Lin Chen, Alex Martinson, Michael Pellin (Deputy Director), David Tiede
Yale University	Victor Batista, Gary Brudvig, Robert Crabtree, James Mayer
University of Chicago	Luping Yu

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Materials Science of Actinides (MSA)
EFRC Director: Peter C. Burns
Lead Institution: University of Notre Dame
Start Date: August 2009

Mission Statement*: To understand and control, at the nanoscale, materials that contain actinides (radioactive heavy elements such as uranium and plutonium) to lay the scientific foundation for advanced nuclear energy systems.*

The Materials Science of Actinides Center unites researchers to conduct transformative research in actinide materials science, with an emphasis on control at the nanoscale. Actinides are, in many ways, at the frontier of exploration of the periodic table, as their chemistry is complicated by the importance of the 5f electrons, relativistic effects of the electrons, their complex redox chemistry, and their radioactivity. Owing to this complexity and the relative difficulty of working with actinides, research in actinide chemistry and actinide-based materials has lagged far behind that of most other elements in the periodic table, both in theory and synthesis and design for special properties, such as radiation resistance of actinide-bearing materials.

In actinides, the delocalization/localization of 5f electrons presents the possibility of control of materials processes at the level of electrons. These properties emerge from the complex correlations of atomic (composition and short and long-range order) and electronic (f-electron) constituents. In this center, we heavily emphasize new synthesis approaches for actinide materials that are likely to lead to revolutionary new forms of matter with tailored properties. New materials that we emphasize are based upon the self-assembly of actinides into nanoscale materials with the potential to create new technologies. Radiation in actinide materials creates a system that is very far away from equilibrium, and a core focus of this center is to examine the behavior of actinide-based materials under extreme conditions of radiation, pressure and temperature.

Four major Research Themes in actinide materials science are central to MSA's efforts. These themes are: Nanoscale cage clusters, complex ceramic and metallic materials, hybrid materials, and materials under extreme environments. Four cross-cutting themes are: Novel synthesis methods for actinide materials across length scales, thermodynamics of actinide materials across length scales, integration of computational analysis and experimental results for actinide materials, and relevance of research to the nuclear fuel cycle.

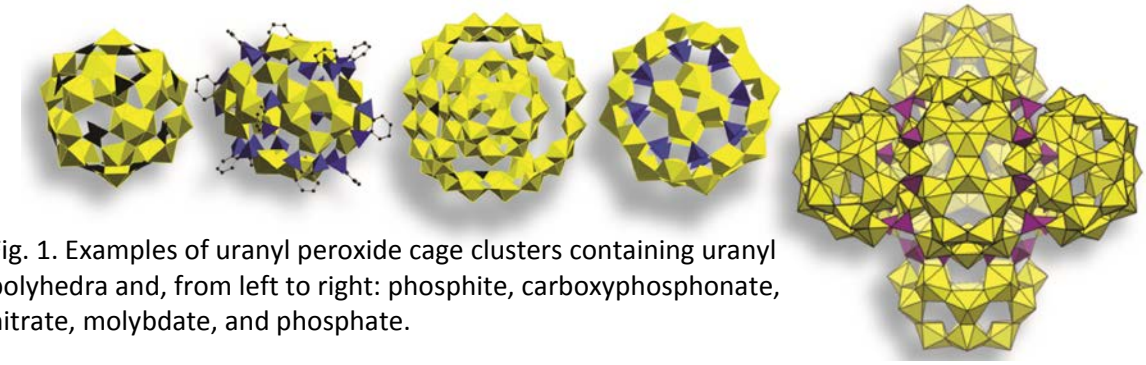

Fig. 1. Examples of uranyl peroxide cage clusters containing uranyl polyhedra and, from left to right: phosphite, carboxyphosphonate, nitrate, molybdate, and phosphate.

The nanoscale cage clusters theme focuses on the self-assembly and properties of a large family of nanoscale actinyl-based cage clusters discovered by this group (Fig. 1). We seek to develop a fundamental understanding of the science of nanoscale actinide materials. Emphasis is on the assembly mechanisms, solution behavior, aggregation, stability, bonding, and ion pairing of these clusters.

The complex ceramic and metallic materials theme extends rigorous experimental determination and computational simulation of thermodynamic parameters of important actinide materials including oxides, selected metals, intermetallics, nitrides, and carbides containing plutonium, neptunium, or uranium. Work includes measurement of heat capacity, thermal expansion, and elasticity of a variety of actinide materials to guide theory and to provide critical data for next-generation nuclear reactors.

The hybrid materials theme includes the synthesis of uranyl based coordination polymer materials with targeted architectures through controlled assembly of selected uranyl building units, and the development of chemical protocols and procedures for modifying actinyl cage clusters with organic ligands, post-assembly, and to investigate how ligation changes the fundamental chemistry of these clusters (Fig. 2).

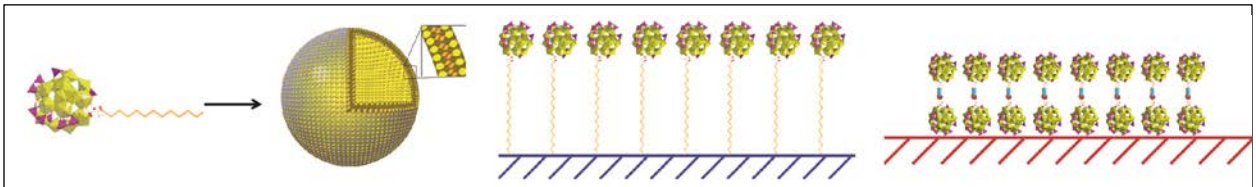

Fig. 2. Hypothetical actinyl cage cluster – organic hybrids.

The materials under extreme environments theme addresses the many phenomena in actinide solids that are radiation dose, temperature, and/or pressure dependent – such as, order-disorder transformations, other phase transitions, and chemical decomposition. The coupling effects of extreme temperature and pressure environments with strong radiation fields are emphasized.

Materials Science of Actinides (MSA)	
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Stanford University	Rodney Ewing, Wendy Mao
University of California-Davis	Alexandra Navrotsky, Mark Asta, William Casey
George Washington University	Christopher Cahill
Oregon State University	May Nyman
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Fluid Interface Reactions, Structures and Transport (FIRST)
EFRC Director: David J. Wesolowski
Lead Institution: Oak Ridge National Laboratory
Start Date: August 2009

Mission Statement*: To develop fundamental understanding and validated, predictive models of the unique nanoscale environment at fluid-solid interfaces that will enable transformative advances in electrical energy storage and electrocatalysis.*

Fluid-Solid Interfaces (FSIs), such as those shown in **Figure 1**, share a common characteristic – the juxtaposition of dense fluids (*e.g.* aqueous, polar organic, ionic liquid), with charged and reactive surfaces (*e.g.* electrodes, catalyst substrates). Transport of ions and molecular reactants through the fluid (often nanoconfined) to the surface (often nanotextured) results in charge storage and/or surface reactions. The unique structural and dynamic properties of the FSI emerge from a complex interplay of short- and long-range forces and reactions among the molecular fluid components, solutes and substrates. The finite size, shape, directional bonding, charge distribution and polarizability of solvents and solutes are convoluted with their ability to reorient, 'un-mix', and react with one another and the substrate. The truncated surface exposes under-coordinated atoms, defects, dopants and active sites that drive interactions with the fluid by bond relaxation, charge redistribution, sorption and intercalation. The FIRST Center addresses fundamental gaps in our current understanding of FSIs to answer these questions of high importance to future energy technologies:

- *How does the interfacial region differ in structure, dynamics and reactivity from the bulk properties of the juxtaposed fluid and solid phases?*
- *How do altered interfacial fluid properties couple with complex substrate nanogeometries and functionalities to influence chemical reactions, ionic and molecular transport and charge transfer within and across the interface?*
- *Can we achieve a quantitative, atomic-nanoscale understanding of FSI phenomena that will enable informed selection and design of fluid and solid components, chemistries and interfacial geometries to attain desired FSI functionality?*

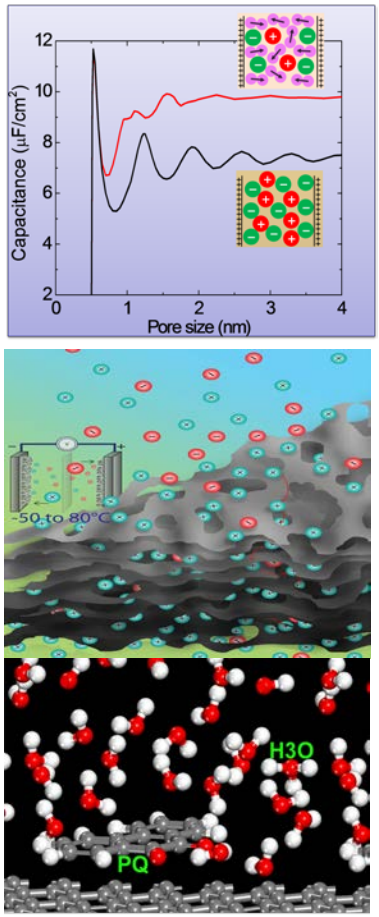

Figure 1 Top: Theoretical prediction of capacitance of nanoporous electrodes in a dipolar solvent (red) versus ionic liquid (black) – (Jiang and Wu, *J. Phys. Chem. Lett.* **2013**, *4*, 1260). Middle: Activated graphene electrode in eutectic ionic liquid (Tsai, et al., *Nano Energy* **2013**, *2*, 403); Bottom: Redox-active quinone (PQ) at graphene-water interface (Anjos, et al., *Carbon* **2012**, *52*, 150).

Our goal is to unleash the ever-increasing power of computer simulations to illuminate the atomistic origins of FSI phenomena and guide the creation of new systems for energy storage and conversion. However, because of current limitations in computational approaches, modeling efforts must be integrated with atomic/nanoscale and macroscopic experimental studies of real FSIs. For instance, we synthesize and probe very simple

interfaces, such as single graphene sheets, atomically-flat single crystal surfaces, etc., in contact with simple electrolyte solutions, in order to guide and validate the computational models of such interfaces, and in turn to use these models to better interpret the experimental results. We then use insights and inferences from these models, applied to more complex interfaces such as slit pores, nanoscale surface curvature, etc. to predict their interfacial structures and transport properties, which can then be tested in real functional interfaces such as supercapacitor electrodes and catalytic substrates.

Ultimately, a range of multiscale models will be needed in order to fully understand and predict the properties of complex interfaces. An important goal of the FIRST Center is to determine how simple our interfacial models can be, and still capture the essential behavior of the interfaces of interest, such as our classical density functional theory models applied to supercapacitors (**Fig. 1**, top). Such coarser-grained models are generally far less computationally-intensive than first-principles approaches, enabling us to simulate much larger systems over much longer time scales than can be addressed using even the currently-available petaflop computers. Another foundational aspect of the FIRST Center is our ability to synthesize and characterize novel substrates (including various forms of carbon and transition metal carbonitrides) and electrolytes (including room temperature ionic liquids) with highly controlled properties and unique functionality (**Fig. 1**, middle). Finally, in order to probe the atomic/nanoscale properties if interfacial systems, we apply a sophisticated array of neutron and synchrotron X-ray scattering (**Figure 2**) methodologies, and we have developed entirely new analytical approaches, such as electrochemical strain microscopy (*e.g.* Black et al., *Adv. Energy Mater.* **2013**, *4*, 1300683).

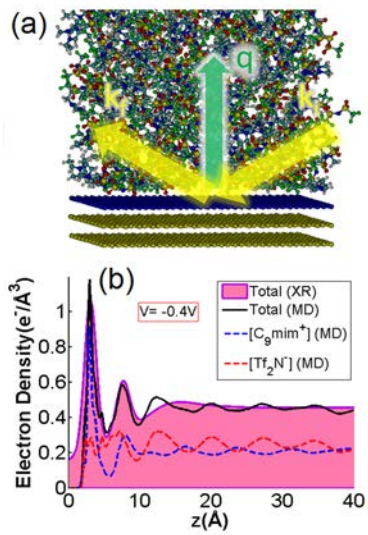

Figure 2. Total X-ray reflectivity (XR, schematic of experimental configuration at top) from a room temperature ionic liquid at a charged graphene surface, compared with predicted structure from a classical molecular dynamics (CMD) simulation (bottom) (Uysal, et al., *J. Phys. Chem. C* **2013**, *118*, 569).

Fluid Interface Reactions, Structures and Transport (FIRST)	
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Penn State University	Adri van Duin
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Energy Dissipation to Defect Evolution (EDDE)
EFRC Director: Yanwen Zhang
Lead Institution: Oak Ridge National Laboratory
Start Date: August 2014

Mission Statement*: To develop a fundamental understanding of energy dissipation mechanisms through electrons, phonons, and magnons, and ultimately control defect evolution in a radiation environment; and to yield new design principles for radiation–tolerant structural alloys for applications in nuclear energy.*

Means of improving the performance of structural materials have been intensively investigated for many decades. Solid solution strengthening, traditionally by alloying minor elements into pure metals, is one of the most widely used methods to achieve specific desirable properties, including radiation tolerance. While it has long been recognized that specific compositions in binary and some ternary alloys have enhanced radiation resistance, it remains unclear how the atomic structure and chemistry affect defect formation and damage evolution. This knowledge gap has been a roadblock to future-generation energy technologies.

The evolution of radiation–induced defect concentration in alloys can be described by three simplified competing processes (Fig. 1): defect production from collision cascades (1st term), subsequent vacancy–interstitial recombination within the diffusion volume (2nd term), and absorption of point defects by extended defects (sinks) such as dislocations, additional phases, interfaces, grain boundaries, and precipitates (3rd term). This evolution has long been considered to involve primarily localized atomic displacements. However, a predictive model of radiation damage will require fundamental understanding of roles of electrons, phonons, and magnons in energy dissipation.

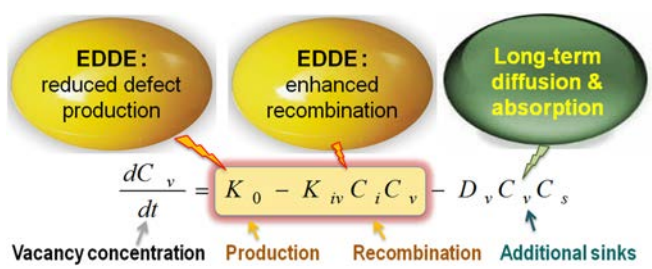

Fig. 1 Simplified terms controlling radiation–induced vacancy concentration. K_0 is the defect production rate; K_{iv} the recombination rate coefficient; and C_i, C_v, D_v and C_s the interstitial and vacancy concentration, the vacancy–sink coefficient, and the sink density; respectively. Similar equations hold for interstitials and other defects.

A new class of materials shows great promise of synthesis with an atomic-level control: Single–Phase Concentrated Solid Solution Alloys (SP–CSAs) containing two to five or more multiple principal elements. To realize the potential of these transformative alloys, we must understand the roles of all constituents in their structural stability and their effects on energy dissipation mechanisms at the level of electrons and atoms. While most of the current research effort addresses the 3rd term to increase sink density, we focus on the reduction of damage accumulation by acting on the early stages of radiation effects (the 1st and 2nd terms). In the EDDE Center, we will advance progressively in alloy complexity from elemental Ni to quinternary SP–CSAs (Fig. 2). Studying this novel series of systems with increasing chemical complexity offers a powerful means to explore the influences of energy deposition and transport, as well as defect formation energies and migration barriers, on defect production and recombination.

The overarching goal of the EDDE EFRC is to develop a fundamental understanding of how the energy of radiation is dissipated, and ultimately to control defect dynamics and microstructural evolution in

structural alloys. Specifically, we seek to understand and quantify the mechanisms of energy dissipation through electronic, vibrational, and magnetic excitation, and how these mechanisms are influenced by alloy complexity.

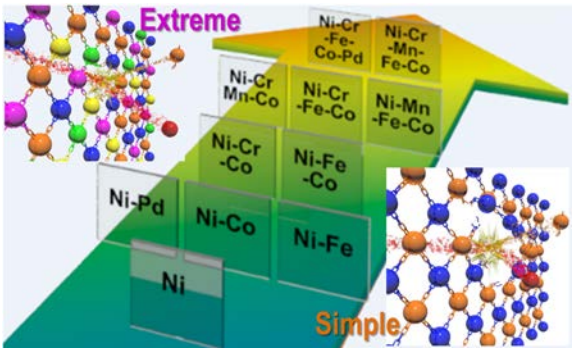

Fig. 2 Ever increasing complexity in *fcc* single–phase concentrated solid solution alloys.

Two thrusts are designed to test our hypothesis that alloy complexity can be tailored to minimize defect production and enhance recombination (Fig. 1). We will develop an understanding in Thrust 1 of *excitation, relaxation, atomic motion, and defect production in model alloys* for which detailed theoretical predictions can be tested with experiments. The focus in Thrust 2 is on *energy dissipation processes and defect evolution in alloys with increasing complexity*. We take advantage of recent successes in synthesis of the nontraditional multi-element alloys (Fig. 2) with compositions at or near equiatomic ratios, including high entropy alloys, to go beyond traditional alloy development. We will also take advantage of recent theoretical and computational developments, many of them by members of our Center, to explore for the first time a comprehensive electronic and atomic description of an irradiated material very far from equilibrium. State-of-the-art synthesis, controlled irradiations, *in situ* ion beam analysis, and post-irradiation microstructure characterization techniques will all be utilized, including channeling image scans, advanced analytical transmission electron microscopy, atom probe tomography, and X-ray and neutron scattering methods. Experimental results, especially on model alloys, will be quantitatively compared to predictions from specialized density functional theory, classical molecular dynamics, and kinetic Monte Carlo techniques. The combination of experiments and these multi-scale computational approaches will offer us the possibility to probe and understand the critical knowledge for controlling and engineering material properties and performance at the ultimate scale – that of atoms and electrons.

The EDDE Center engages a diverse mix of principal investigators and senior/key personnel who have complementary experience and skills. Each thrust will incorporate both theory and experiment. Most participants will contribute to both thrusts, thus maximizing synergies and coordination. The strong university involvement will enhance educational outreach. The success of the EDDE Center will yield new design principles for radiation-tolerant structural alloys for applications in nuclear energy and new defect engineering paradigms for much broader science and technologies.

Energy Dissipation to Defect Evolution (EDDE)	
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The Center for Molecular Electrocatalysis (CME)
EFRC Director: Morris Bullock
Lead Institution: Pacific Northwest National Laboratory (PNNL)
Start Date: August 2009

Mission Statement*: To understand and design molecular electrocatalysts for conversions between electrical energy and chemical energy.*

Electrocatalysts that efficiently convert electrical energy into chemical bonds in fuels, or the reverse, converting chemical energy to electrical energy, will play a critical role in future energy storage and energy delivery systems. Electrocatalytic processes involving multi-proton and multi-electron redox reactions are pervasive in energy science, and are critical for sustainable, carbon-neutral energy. The Center for Molecular Electrocatalysis addresses fundamental challenges in understanding how molecular electrocatalysts function, and uses this knowledge to rationally design new classes of molecular electrocatalysts for important energy storage and utilization reactions. Closely coupled experimental and theoretical studies include inorganic synthesis, ligand design, computational studies, mechanistic investigations, electrochemical and spectroscopic measurements, determination of thermochemical values for metal complexes, and evaluation of catalytic activity.

$$2\,H^+ + 2\,e^- \rightleftharpoons H_2 \qquad (1)$$

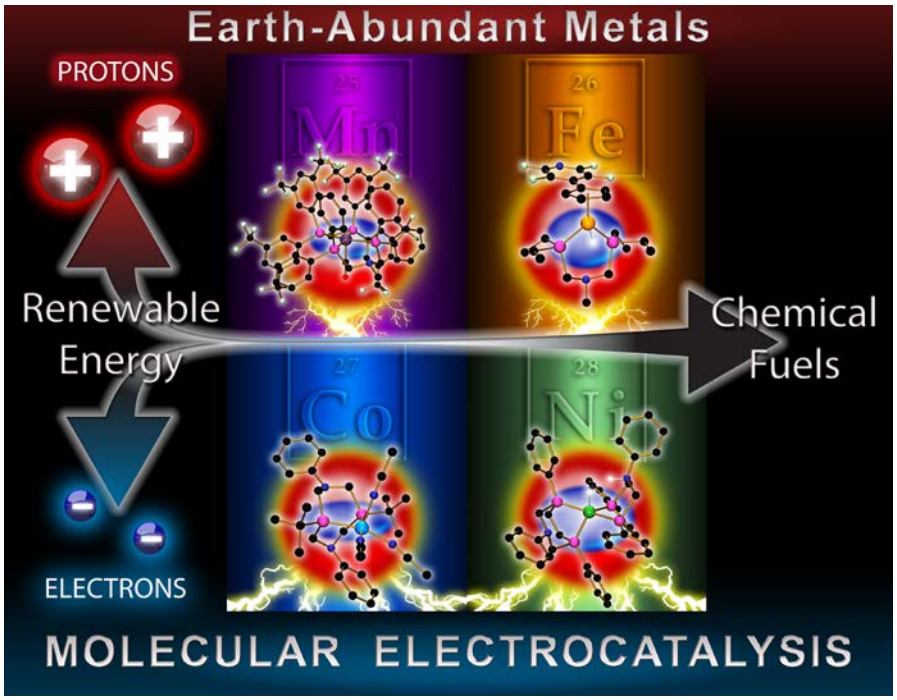
$$O_2 + 4\,H^+ + 4\,e^- \rightleftharpoons 2\,H_2O \qquad (2)$$

$$N_2 + 6\,H^+ + 6\,e^- \rightleftharpoons 2\,NH_3 \qquad (3)$$

Electrocatalytic reactions studied range from two-proton, two-electron processes to six-proton, six-electron reactions. A unique approach in this Center is a focus on proton relays, which are functional groups (typically amine bases or carboxylic acids) that play a crucial role in the delivery of protons to (or from) the active site of molecular catalysts. *The prevalence in energy science of reactions that require controlled movement of protons and electrons presages an immense scope for the roles of proton relays.*

The reduction of protons derived from water to form hydrogen is shown in eq. 1 (forward direction). The reverse process, the oxidation of H_2, is the reaction used in hydrogen fuel cells to convert the chemical energy in the H-H bond to electricity. The four-electron reduction of O_2 to form water, (eq. 2, forward direction), is important in many fuel cells, providing the reductive half-reaction to balance the oxidative half-reaction of H_2. The opposite process, (eq. 2, reverse direction), the four-electron oxidation of water to form O_2, is required for water splitting, and has been intensively studied in connection with solar energy utilization. The reduction of nitrogen to ammonia stores energy in N-H bonds, and use of NH_3 in fuel cells converts the chemical energy of the N-H bonds to electrical energy.

Molecular catalysts offer a high degree of structural control – and therefore the precise probing of relationships between catalyst structure and activity – that are much more difficult for heterogeneous catalysts and enzymes. *We seek to further develop this knowledge of structure-activity relationships.* The two, four-, and six-*electron* redox processes in eqs. 1-3 for H_2, O_2, and N_2, are also two-, four-, and six-*proton* processes. Facile and controlled movement of both electrons *and* protons from solution to substrates bound at the active metal site is essential for these electrocatalytic reactions. Proton transfers must be very carefully controlled to obtain optimal rates and efficiency of molecular electrocatalysts. The generality of proton transfer processes in most fuel generation and utilization reactions makes understanding these processes at a fundamental level of enormous importance.



Proton relays are functional groups that play a crucial role in the delivery of protons to (or from) the active site of catalysts. They have been shown to play an important role in hydrogenase enzymes, the oxygen-evolving complex, and other biological systems.

Primary goals of the Center for Molecular Electrocatalysis are to:

- Obtain a fundamental understanding of how proton relays accelerate *intra*molecular and *inter*molecular proton transfers
- Understand how proton transfers can be coupled with electron transfers to accelerate catalytic reactions
- Demonstrate that a comprehensive understanding of proton relays can be used to design highly active molecular electrocatalysts for multi-proton, multi-electron reactions.

Center for Molecular Electrocatalysis (CME)	
Pacific Northwest National Laboratory	Morris Bullock (Director), Monte Helm (Deputy Director), Aaron Appel, Michel Dupuis, Michael Mock, Simone Raugei, Molly O'Hagan, Eric Wiedner
Yale University	James Mayer
University of Illinois	Sharon Hammes-Schiffer
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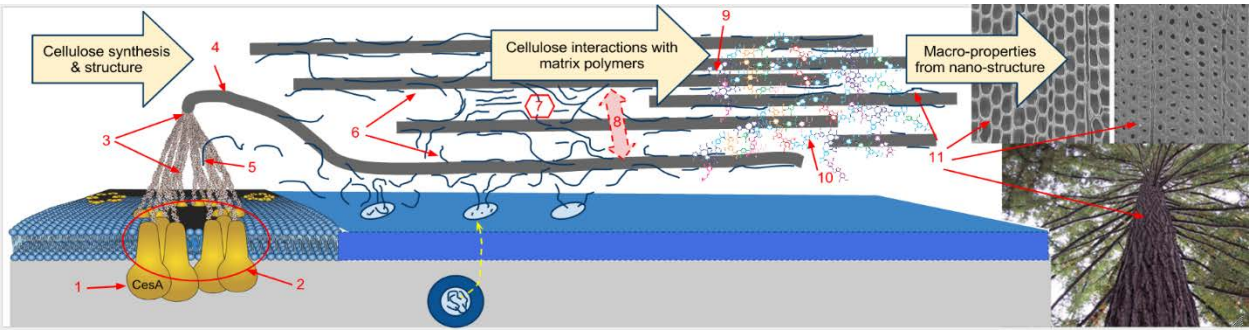
The Center for Lignocellulose Structure and Formation (CLSF)
EFRC Director: Daniel J. Cosgrove
Lead Institution: Pennsylvania State University
Start Date: August 2009

Mission Statement*: to develop a nano- to meso-scale understanding of plant cell walls, the main structural material in plants, and the mechanisms of their assembly, forming the foundation for significant advances in sustainable energy and novel biomaterials.*

Cellulosic biomass (lignocellulose) holds great promise as a large-scale, renewable and sustainable source of liquid biofuels for transportation, if we could overcome technical obstacles stemming from its complex, hierarchical structure. Despite its huge economic importance, many aspects of lignocellulose structure and formation remain shrouded in mystery. For instance, little is known of the details of how the cellulose-synthesizing nano-machine at the cell surface links simple sugar molecules into long strands and extrudes them at the cell surface in such a way that they make a strong, insoluble and highly inert crystalline fibril. Likewise, the processes by which simple polymers are transformed into a strong and recalcitrant biomaterial are not well understood.

CLSF goals are to develop a detailed nano- to meso-scale understanding of plant cell wall structure and its mechanism of assembly, from the molecular mysteries of how glucose is assembled by cellulose synthase complexes to form cellulose microfibrils to the orderly, hierarchical interaction of cellulose with other components to form cell walls with diverse properties. The diagram below sketches some of the key points for lignocellulose formation, starting with cellulose synthesis by cellulose synthases (CESA) organized into a complex embedded in the plasma membrane (left, points 1-2), followed by cellulose crystallization and interactions with hemicelluloses to form a cohesive wall (points 3-8) and then changes that accompany lignin polymerization within the wall (points 9-11).



New understanding of these processes will form the scientific foundation for designing rational, science-based pretreatments to deconstruct cell walls and for using genetic techniques to coax plants into making modified walls for significant advances in sustainable energy and novel materials. It will also yield insights into biomimetic ways to transform simple molecules into complex polymeric materials with diverse physical and chemical properties.

CLSF research is organized around two themes concerned with the process of cellulose synthesis and the rules of assembly of cell wall components to make a cell wall with specific physical properties.

CLSF Theme 1 probes the mechanism of cellulose microfibril (CMF) formation. Specific objectives include: (1.1) Determination of the structure and in-vitro activity of plant cellulose synthase (CESA) by

biochemical means, X-ray crystallography and electron microscopy, as well as by computational modeling of the dynamics of glucan synthesis by a CESA protein. (1.2) Assessing the roles of parts of the CESA protein for cellulose synthesis by genetic modification and analysis of CESA activity and cell wall phenotypes. (1.3) Analysis of the contributions of CESA isoform to the activity of the Cellulose Synthesizing Complex (CSC) and properties of the microfibril and macrofibril through microscopic and genetic experiments. (1.4) In-depth characterization of the plant CSC by isolating an active CSC, modeling CMF formation in silico, and identifying proteins that interact with CSCs that synthesize cellulose in secondary cell walls. (1.5) Reconstitution of a functional CSC from purified plant components by assembling the essential components of the plant CSC.

CLSF Theme 2 investigates the nano- and meso-scale structure and assembly of cell walls and the basis for their important physical and biological properties. Research objectives include: (2.1-2.2) Analysis of mesoscale wall architecture and dynamics by Atomic Force Microscopy (AFM), small-angle neutron scattering, and solid-state Nuclear Magnetic Resonance (ssNMR) in combination with enzymatic and genetic modifications of the wall. (2.3) Testing of the current model of the grass cell wall using enzymatic, biomechanical and physical approaches. (2.4) Testing how matrix polymer delivery relates to cellulose biosynthesis and order using click-labeling of matrix polysaccharides combined with analysis by AFM and transmission electron microscopy. (2.5) Analysis of the mobility of water, polysaccharides and proteins by neutron scattering and ssNMR to assess influence of wall components and wall charge on wall dynamics. (2.6-2.7) Incorporation of new as well as existing data into computational models of primary and secondary cell wall architecture and material properties. (2.8) Analysis of the physical effects of lignin polymerization in cell wall explants and analogs, including mobility of water and polysaccharides. (2.10) Development and use of sum frequency generation (SFG) spectroscopy (a) to analyze meso-scale organization of cellulose in single cell walls and (b) to refine the interpretation of SFG spectra.

The Center for Lignocellulose Structure and Formation (CLSF)	
Penn State University	Daniel J. Cosgrove (Director), Charles Anderson, Gong Chen, Ying Gu, Seong Han Kim, James Kubicki, Manish Kumar, Janna Maranas, Karl Mueller, B. Tracy Nixon, Ming Tien
North Carolina State University	Candace Haigler, Yaroslava Yingling
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University of Rhode Island	Alison Roberts
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Center for direct Catalytic Conversion of Biomass to Biofuels (C3Bio)
EFRC Director: Maureen McCann
Lead Institution: Purdue University
Start Date: August 2009

Mission Statement*: To master the ability to reconfigure all partially reduced carbon from plant cell walls into desired molecules.*

New capabilities to predict, design and control the chemistries of carbon could enable the transition from fossil-based to sustainable transportation fuels. Lignocellulosic biomass, a renewable and carbon-neutral resource, has the potential to displace an estimated annual equivalent of three billion barrels of oil in the U.S. alone. However, biomass has only one-third the energy density of crude oil and lacks petroleum's versatility as a feedstock for fuels and chemicals. These limitations keep biomass conversion below the efficiency level needed for strategic impact while the scientific challenge of routing carbon from one molecular context to another remains unmet.

The polysaccharides and lignins of the plant cell wall form a complex, cross-linked polymeric structure of substantial physical and chemical resilience that impedes access of catalysts to targeted chemical bonds. Its complexity results in heterogeneous product streams after catalytic or pyrolytic processing. Cross-links among plant cell wall biopolymers generate nanoscale architectures and distinct mesoscale domains that have dramatically different properties than those observed in mixtures of biopolymers. C3Bio research demonstrated that the disparity between theoretical and actual yields of liquid hydrocarbons and high-value chemicals is a consequence of this structural complexity. C3Bio will now develop critical systems-level understanding of how biomass structural complexity at molecular, nanoscale, and mesoscale levels impacts the yields and selectivities of desired reaction products from catalytic and pyrolytic transformations. We will establish the fundamental science required to modulate cell wall complexity and catalytically transform intact biomass in order to gain unprecedented control of effective routing of carbon: we will specify both the structures within, and the reaction products from, lignocellulosic biomass.

Goal 1: Develop Catalytic and Pyrolytic Processes Specifically Designed for the Structural Complexity of Biomass. We will investigate the kinetics and mechanisms of catalytic and pyrolytic pathways to advance applicability and selectivity in intact biomass conversion and will use native and synthetic biopolymers and biomass genetic variants to define modified substrate performance in conversion pathways. Goal 1 impacts include lignin evolving from a material used primarily as biorefinery process fuel to a material with utility and value equal to that of biorefinery carbohydrates. We will gain ability to deliver intermediate products from biomass polysaccharides in high yields and with carbon atom conservation for upgrading to drop-in liquid fuels. Pyrolysis will become a feasible technology to make simplified bio-oils for HDO catalysis.

Goal 2: Redesign the Structure of Biomass for Carbon- and Energy-efficient Catalytic and Pyrolytic Transformations*.* C3Bio data on biomass performance in catalytic conversion provide new targets for the genetic engineering of tailored biomass, push the state-of-the-art of meso-scale and multi-scale modeling, and allow for greater predictability and understanding of properties of molecular architectures. At mesoscale, we will modulate localized carbon density, cell-cell adhesion, and target modifications to specific cell types. Multi-scale modeling will accelerate identification of the most chemically labile and stable linkages. Goal 2 impacts will be new understanding to use trait stacking to increase the intrinsic carbon density of the biomass and also brittleness at the time of comminution to

produce optimized biomass. A codesign approach will maximize yield and selectivity of reaction products by modifying the substrates *in planta* and creating novel catalyst delivery systems.

Goal 3: Deliver Innovative Pathways for Targeted Product Portfolios from Tailored Biomass. Post-catalytic residues provide modified substrates and architectures for subsequent transformations. We will apply our imaging and analytical toolkits to identify the control points and deliver legacy scientific understandings of how chemical reaction kinetics and biomass properties at the nano-scale (porosity, surface chemistry) and meso-scale (particle size and geometry) affect product formation rates and product profiles. We aim to enable advanced catalyst, reactor, and biomass design beyond "no carbon left behind" to achieve unprecedented control over the routing of carbon from biomass into fuels and chemicals – "a place for every carbon and every carbon in its place".

Center for direct Catalytic Conversion of Biomass to Biofuels (C3Bio)	
Purdue University	Maureen McCann (Director), Mahdi Abu-Omar (Associate Director), Nathan Mosier (Associate Director), Rakesh Agrawal, Nicholas Carpita, Clint Chapple, Nicholas Delgass, Hilkka Kenttamaa, Rick Meilan, Fabio Ribeiro
National Renewable Energy Laboratory	Gregg Beckham, Michael Crowley, Bryon Donohoe, Michael Himmel, Melvin Tucker
University of Tennessee	Craig Barnes, Joseph Bozell, Alison Buchan
Northeastern University	Lee Makowski

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765-496-9640, http://c3bio.org

Center for Mesoscale Transport Properties (m2m)
EFRC Director: Esther Takeuchi
Lead Institution: Stony Brook University
Start Date: August 2014

Mission Statement*: To understand and provide control of transport properties in complex battery systems with respect to multiple length scales, from molecular to mesoscale (m2m); to minimize heat and maximize work of electrical energy storage devices.*

At its essence, energy is the sum of heat and work: $\Delta E = q + w$. As such, the ultimate goal for any energy storage system is to maximize useful work (w) and minimize the generation of waste heat (q). During the operation of an energy storage system, ions and electrons are transported over multiple size domains where the sum of these processes leads to complex physics. Resistance evolves over time due to phase changes in the solids and changes in the composition and structure of the interfaces. These complicating factors must be considered to derive the full panoply of information needed for rational design and predictive modeling of materials useful in energy storage systems. While inefficiency can be approached at the macro level, emphasizing bulk parameters and bulk methods cannot fully interrogate or address the inherent heterogeneity of ion and electron flux contributing to the local resistance within an electrode and at the interfaces. In order to develop the capability to predict and ultimately control energy storage systems, these inefficiencies must be understood not just as a bulk property (heat), but rather as localized resistance at the molecular to mesoscale (*m2m*) levels.

The goal of the *m2m* EFRC is to enable deliberate design of materials and components to achieve higher performing, longer life, and safer energy storage systems through acquisition of new fundamental knowledge about ion and electron transport and electron transfer properties of energy relevant materials, over multiple length scales, across interfaces and over time. The expected research outcomes are that the Center will provide the conceptual approaches to predict materials properties, processing outcomes, and functional characteristics which determine conduction and electron transfer properties, including the complexities of interfaces and time. The information gained will enable design of materials and systems to bridge the gap between theoretical energy content and functional energy delivery.

The Center will frame the key scientific investigations along three Scientific Inquiry Areas (SIA) that pose the following questions:

1. What are the *fundamental limits* of ion and electron transport and electron transfer over multiple length scales?

This question will be addressed by investigating transport and transfer phenomena for redox active moieties with several configurations including those intimately bound to conductive solids. The roles of structure, crystallite size and particle size will be considered.

2. How do the phenomena change across *multiple domains* including interfaces? Which systems level interactions are synergistic, independent, or antagonistic?

The influence of physical properties, surface functionalization, and process on the transfer and transport properties and the function of composite electrodes will be explored. The feasibility of deliberately tuning interfaces by surface modification or functional group modification of the solids and electrolyte will also be probed.

3. How do the transport phenomena evolve *over time* in systems not at equilibrium?
The effect of extended ion or electron flux on energy related solids will be studied to determine the effect of time. Additionally, the feasibility of fabricating electrodes under the influence of an external force to achieve enhanced transport behavior will be investigated.

The ***m2m*** Center will achieve the goals via:

1) integration of researchers in materials, characterization, theory and electrochemical systems interrogation.
2) use and development of microscopic and nanoscopic science based tools to provide characterization insights, highlighted in Figure 1.
3) synthetically tunable model redox active materials and electrolytes which will be shared among center members to facilitate interaction and data interpretation.
4) frequent communication and assessment.

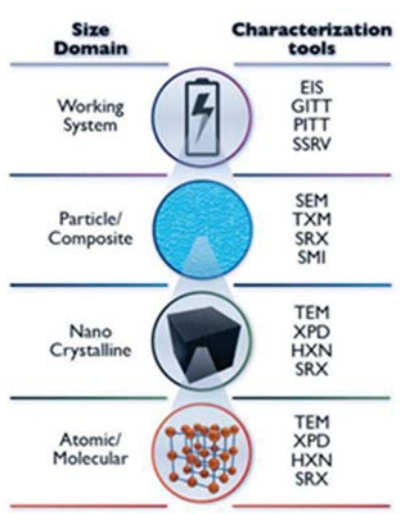

Figure 1. Characterization tools across multiple size domains

The Center will access and utilize five notable facilities: the Advanced Energy Research and Technology Center (AERTC) and CEWIT at Stony Brook University, and the Interdisciplinary Science Building (ISB) including a 1200 sq ft dry room, National Synchrotron Light Source II (NSLS-II) scheduled to begin operation in 2014, and Center for Functional Nanomaterials (CFN) at Brookhaven National Laboratory. AERTC will be the home base of the Center, providing access to a suite of new analytical instrumentation. CEWIT (Center of Excellence for Wireless Information Technology) will act as the data storage site for the Center.

Center for Mesoscale Transport Properties (*m2m*)	
Stony Brook University	Esther Takeuchi (Director), Amy Marschilok (Center Operations Officer), Kenneth Takeuchi, Stanislaus Wong, Maria Fernandez-Serra
Brookhaven National Laboratory	Mark Hybertsen, Eric Stach, Feng Wang, Jun Wang, Yimei Zhu
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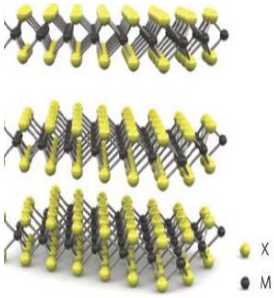
Center for the Computational Design of Functional Layered Materials (CCDM)
EFRC Director: John P. Perdew
Lead Institution: Temple University
Start Date: August 2014

Mission Statement*: To design new or defect-engineered functional layered materials on the computer, for applications such as solar cells, batteries, and catalysts to generate hydrogen fuel or to convert carbon dioxide to methanol.*

New or modified materials with desired functionalities play an essential role in the development of clean-energy technologies, such as solar cells, batteries, and catalysts to generate hydrogen fuel by water-splitting. Because there are so many possible materials, it is impossible to grow and test them all in the laboratory. Computational design of materials with desired properties, based on first-principles theory and modeling, is a practical alternative. (The figure shows the structure of layered MX_2, from Wang *et al., Nature Nanotechnology* ***7***, 699 (2012).)



Layered materials are typically composed of planar sheets, with strong bonds between the atoms within a layer and weak bonds between the layers. A collection of many layers is a three-dimensional (3D) material, while one or a few layers make a 2D material. Variation of properties with dimensionality is an appealing feature of these materials. The 2D materials can be grown on a substrate, or exfoliated from the 3D materials. Extraneous atoms can bind at the surface, or take up residence between the layers. The surfaces of many materials are contaminated by contact with air. The layered materials interact only weakly with the molecules in air, but can be modified by controlled exposure to free atoms.

Some layered materials have no fundamental energy gap. Graphene (unsupported 2D carbon) is a celebrated example. But interest for applications is now shifting to "beyond-graphene" 2D semiconductors, such as boron nitride (BN) or molybdenum disulfide (MoS_2), and to heterostructures made by stacking different 2D crystals. Other transition-metal dichalcogenides (TMD's MX_2, as pictured above), such as WS_2, WSe_2, and $MoSe_2$, are also of interest, as are layered oxides.

Layered 2D and 3D materials display many potentially useful properties. The direct energy gap in 2D MoS_2, which can be tuned by material modification, opens up the possibility of flexible optoelectronic or photovoltaic applications. Photo-excited electrons and holes can be spontaneously separated in a 2D heterostructure. A single layer of the 1T polytype of MoS_2 can catalyze the photo-induced splitting of water. Small ions can intercalate between the layers of a material, as in a lithium battery.

We will design new functional layered materials by theory, modeling, and computation. Candidate materials of special interest will be grown and experimentally characterized in the Center. Experimental and theoretical work will be carried out for catalysis on layered materials, e.g., water splitting.

Thrust 1. *Density functional and correlated-wavefunction methods development*. Improved density-functional and (embedded) correlated-wavefunction methods will be developed for computational modeling of many-electron systems and especially layered materials. A strong component of fundamental methods development and testing is a unique contribution of our Center.

Thrust 2. *First-principles computation and design of layered materials*. First-principles computations will be performed for many layered materials, and for their modifications due to a variety of localized or periodic defects and perturbations (defect engineering). In this way, new layered materials with desired functionalities will be predicted.

Thrust 3. *Multi-scale modeling for extended defects in materials*. Multi-scale models with first-principles input will be developed and applied to extended defects in layered materials, including dislocations, grain boundaries, and interface sliding in TMD's. The stability of grain boundaries between domains of the 2D systems will be investigated, as will the interaction of defects with water.

Thrust 4. *Growth and characterization of layered materials*. Promising candidate layered materials including 2D MoS_2 will be grown, e.g., by self-limiting chemical vapor deposition which can control the number of layers. They will be characterized by Raman spectroscopy and atomic force microscopy, which probe the number of layers, and by low-temperature scanning tunneling microscopy (STM) and spectroscopy, which measure the local density of quasiparticle states. Atoms can be deposited on the surface in ultra-high vacuum before the STM measurements.

Thrust 5. *Catalysis and water splitting in layered materials*. Experimental and theoretical studies will be made for the catalysis of water-splitting on layered materials. For example, the photo-irradiation of layered MoS_2, WS_2, and MnO_2 can potentially catalyze the splitting of water. 2D heterostructures will also be investigated as potential photocatalysts for water splitting. Finally, designed layered materials will be explored as catalysts to convert carbon dioxide to methanol (a fuel).

Center for the Computational Design of Functional Layered Materials (CCDM)	
Temple University	John P. Perdew (Director), Michael L. Klein, Xiaoxing Xi, Eric Borguet, Maria Iavarone, Daniel R. Strongin, Michael J. Zdilla, Xifan Wu, Adrienn Ruzsinszky, Jianwei Sun
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Center for Frontiers of Subsurface Energy Security (CFSES)
EFRC Director: Larry W. Lake
Lead Institution: The University of Texas Austin
Date Started: August 2009

Mission Statement*: to understand and control emergent behavior arising from coupled physics and chemistry in heterogeneous geomaterials, particularly during the years to decades time scales over which injection for geologic CO_2 storage will drive natural systems far-from-equilibrium.*

Geologic CO_2 storage (GCS) is key for mitigating greenhouse gas emissions, but to be effective GCS must overcome three primary technical challenges: sustaining large storage rates for decades, using pore space with unprecedented efficiency, and controlling undesired or unexpected behavior in geostorage. To meet these challenges the *Center for Frontiers of Subsurface Energy Security* (CFSES) will pursue scientific advances with the goal to (i) establish scientific understanding of far-from-equilibrium processes in heterogeneous geologic materials, and (ii) develop novel materials and methods for controlling those processes. Specific examples of our **science goals** are: we seek to

- Understand *chemical-mechanical coupling* by identifying mechanisms and time/length scales for self-reinforcing fracture propagation and fluid leakage through those fractures.
- Combine experiments and computational chemistry to examine the *structural (mechanical) and chemical interactions* between wet supercritical CO_2 and clays. We also seek advanced models of noble gas and isotope partitioning between immiscible fluid phases.
- Understand and predict *modes* (compact viscous flow vs. capillary channels) *and fluxes* of reactive CO_2 migration through brine-saturated geomaterials with cm-to-m scale heterogeneity.
- Capitalize on the improved understanding of these processes to design, develop and apply *novel materials* that will alter fluid-assisted perturbations in heterogeneous geomaterials.

The **impact** of the proposed research will be a new scientific foundation for defining the physical constraints on challenges for GCS, and for identifying novel materials and strategies to overcome these constraints. The science will be valuable for broader policy considerations associated with greenhouse gas emissions, the risks in operating storage sites, and the appropriate mix of incentives and regulation for GCS operation. The science addressed in this work also applies other subsurface processes involving multiple fluids that are important to energy technologies, e.g., methane hydrates, hydrocarbon production and fluid injection.

CFSES have already addressed science questions underlying two current concerns for GCS: storage security and risk assessment.

Science to Inform Storage Security

Emergent behavior during CO_2 transport within faults: We sampled from the natural GCS analog at Crystal Geyser, Utah and the observed CO_2 seeps along the Little Grand Wash fault. We developed scenarios for CO_2 migration and seepage along the fault, and incorporated these into pore-scale models for calcite. The biomass observations indicate organisms can survive under large CO_2 pressures. **Impact:** our work provides new understanding of the evolution of CO_2 leakage paths, and a scientific basis for modeling phenomena relevant to long-term security.

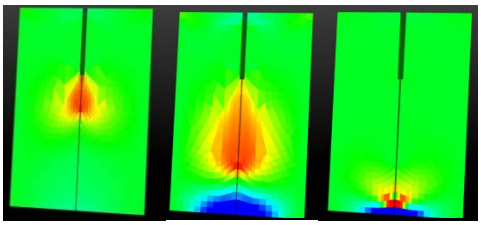
Emergent behavior at the reservoir-scale CO_2/brine interface: Analysis of brine from the Bravo Dome field site enabled the first benchmarked simulations of CO_2 dissolution with realistic brine chemistry. We

explained the enrichment of helium relative to CO_2 observed at Bravo Dome. **Impact:** our work provides insight into pattern emergence in geophysically common buoyancy-driven phenomena. Simultaneously it is yielding the first field-based assessment of the relevance of solution trapping for storage security.

Far-from-equilibrium states during CO_2 displacement of brine: Work shows that surface-treated nanoparticles emplaced in brine can significantly reduce the mobility of CO_2 by preserving a far-from-equilibrium state at the pore scale as the CO_2 displaces brine. **Impact**: the underlying mechanism discovered here could yield a novel method for greatly increasing storage security.

Science to inform GCS risk assessment

Emergent phenomena and far-from-equilibrium behavior during fracture initiation and propagation: We developed a new method for measuring elastic moduli of shale caprocks to determine rock mechanical properties from well log measurements and to determine reservoir scale rock mechanics properties. **Impact**: CFSES established synergistic experimental and modeling capability to understand and control the initiation and propagation of fractures from injection wells.



Cohesive modeling results of crack propagation of short-rod experimental

Far-from-equilibrium CO_2-mediated geochemistry and caprock integrity: We have developed new molecular dynamics tools that provide unprecedented insight into residual trapping of CO_2 into pore space. **Impact:** Our calculations are pioneering work in molecular dynamics, with macroscopic implications for surface phenomena and the new science of CO_2-mediated geochemistry

Emergent phenomena during CO_2 plume migration: signature of heterogeneous fluid/rock properties in acoustic wave propagation: We developed algorithms that extract much more information from time-lapse seismic data than was possible previously. The effectiveness of the technique was validated with the time-lapse surface seismic surveys at the Cranfield (Mississippi) field CO_2 sequestration demonstration site. **Impact:** Our innovation greatly increases the benefit of repeated seismic surveys for risk assessment.

Center for Frontiers of Subsurface Energy Security (CFSES)	
The University of Texas at Austin	Larry W. Lake (Director), Hilary Olson (Assistant Director), Marc Hesse, Tip Meckel, and Sanjay Srinivasan (Theme Leads), Matthew Balhoff, Philip Bennett, M. Bayani Cardenas, David DiCarlo, Peter Eichhubl, Nicolas Espinoza, Omar Ghattas, Nick Hayman, Susan Hovorka, Chun Huh, Keith Johnstone, Mary F. Wheeler, Changbing Yang
Sandia National Laboratories	Marianne Walck (Associate Director), Susan Altman (Assistant Director), Randall Cygan, Thomas Dewers, and Mario Martinez (Theme Leads), Joseph Bishop, Anastasia Ilgen, Craig Tenney, Yifeng Wang, Hongkyu Yoon

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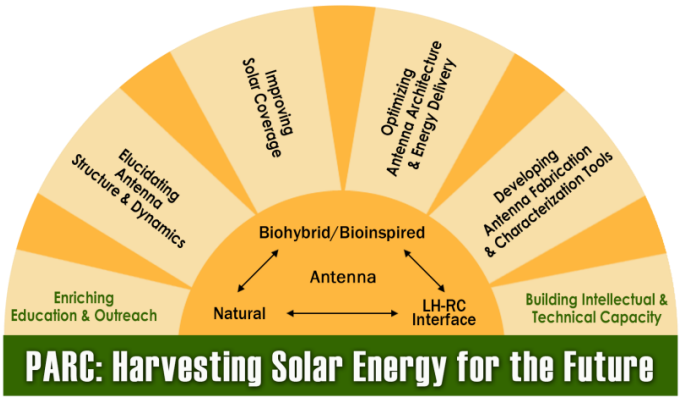
Photosynthetic Antenna Research Center (PARC)
EFRC Director: Robert E. Blankenship
Lead Institution: Washington University in St. Louis
Start Date: August 2009

Mission Statement: *to understand the basic scientific principles that underpin the efficient functioning of natural photosynthetic antenna systems as a basis for design of biohybrid and bioinspired architectures for next-generation systems for solar-energy conversion.*

Scientific Themes. Through basic scientific research, PARC seeks to understand the principles of light harvesting and energy funneling as applied to three programmatic themes:

1. *Natural Antennas*: Structure and Efficiency
2. *Biohybrid and Bioinspired Antennas*: Design and Characterization
3. *Antenna-Reaction Center Interface*: Organization and Delivery



Unifying Research Activities. The three scientific themes are connected by the idea that enhancements of photosynthetic light harvesting and the design of the biohybid and bioinspired antenna will draw upon the lessons learned from natural systems. The thematic activities are joined by interrelated research threads, including to:

- Elucidate antenna structure and dynamics
- Improve solar coverage
- Optimize antenna architecture and energy delivery
- Develop antenna fabrication and characterization tools

Global Impact. All of the PARC activities have the broader goals to:

- Enrich education and outreach
- Build intellectual and technical capacity

Specific Objectives for Theme 1—Natural Antennas: Structure and Efficiency

PARC seeks to understand the structure and function of the wide variety of natural photosynthetic antenna systems, including their efficiency, mechanism of action, regulation, assembly and repair. Emphasis is placed on developing new antenna systems using techniques of molecular and synthetic biology that increase the efficiency and functionality of living photosynthetic organisms. The specific objectives are to:

- Determine structures of native and modified antenna systems and correlate with function
- Understand subcellular organization of antenna systems in different photosynthetic organisms
- Examine physiological consequences of modified/alternate antenna in diverse organisms
- Confer visionary new functional attributes into living photosynthetic organisms

The overall aim is to determine and manipulate the antenna architecture and composition to maximize photosynthetic efficiency and functionality in any such organism.

Specific Objectives for Theme 2—Biohybrid and Bioinspired Antennas: Design and Characterization
PARC will use first principles and readily accessible constituents to create mesoscale antennas with tailorable performance specifications with regards to spectral coverage, absorbance intensity, and efficiency of exciton delivery and transduction. The specific objectives are to:

- Develop self-assembled macromolecular arrays based on tunable synthetic pigments coupled to natural or designer polypeptide scaffolds
- Control mesoscale architecture of antennas in lipid bilayers or liposomes
- Nanopattern arrays of biohybrid/bioinspired antennas and RCs on surfaces in various geometries, including nanoscale lines and dots

The overall aim is to design biohybrid and bioinspired architectures for energy collection and storage.

Specific Objectives for Theme 3—Antenna-Reaction Center Interface: Organization and Delivery
PARC seeks a fundamental understanding of antenna-RC interfaces in native, biohybrid, and bioinspired antennas optimized for total control over molecular (structural, energy, redox) characteristics for delivery of energy, electrons or redox equivalents to downstream processes such as catalysis, fuels production or photocurrent generation. The specific objectives are to:

- Understand and manipulate the interfacing and regulation of antenna and RC units in native photosynthetic systems
- Extend the functionality of biohybrid and bioinspired arrays to include sites of energy trapping and photochemistry at defined positions
- Fabricate multi-component assemblies or networks of electronically communicating antennas and RCs on surfaces to promote long-range, directional energy flow

The overall aim is to understand and control the coupling of antenna and RC functions in solar energy conversion systems.

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GRAND CHALLENGES INDEX

The Basic Energy Sciences Advisory Committee (BESAC) Grand Challenges report, "Directing Matter and Energy: Five Challenges for Science and the Imagination", identifies the most important scientific questions and science-driven technical challenges facing Basic Energy Sciences (BES) and describes the importance of these challenges to advances in disciplinary science, to technology development, and to energy and other societal needs. Each EFRC addresses one or more of these grand challenges. For more information, see http://science.energy.gov/bes/efrc/research/grand-challenges/.

How can we master energy and information on the nanoscale to create new technologies with capabilities rivaling those of living things?.....11, 15, 25, 29, 31, 39, 41, 43, 53, 55, 61, 63

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BES REPORTS INDEX

BES-sponsored workshop reports engaged participants from universities, industry, and DOE laboratories to help identify research directions for a decades-to-century energy strategy. The 32 Energy Frontier Research Centers directly address the use-inspired energy challenges articulated in these reports. For more information, see http://science.energy.gov/bes/efrc/research/bes-reports/.

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