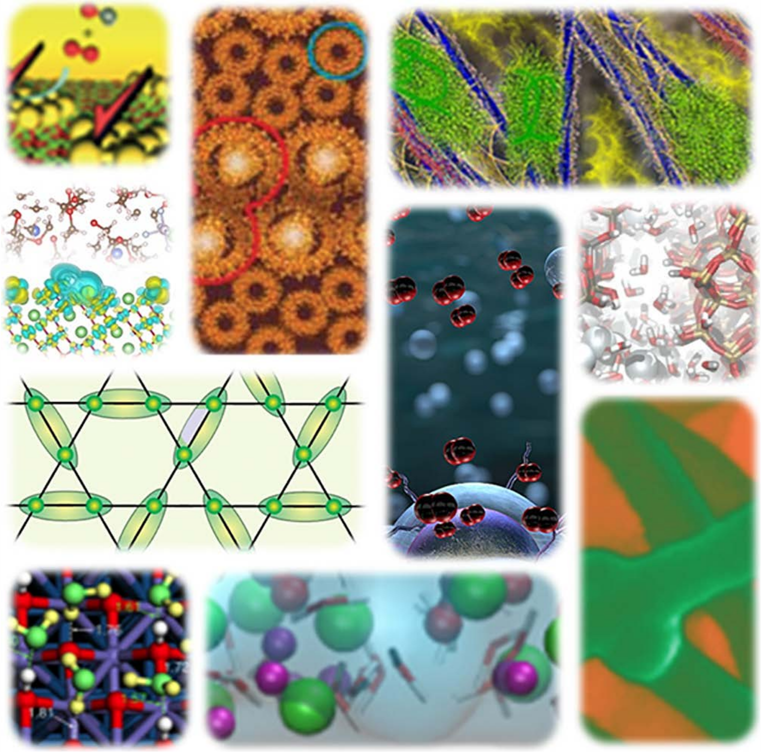
Energy Frontier Research Centers

Technical Summaries



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U.S. DEPARTMENT OF ENERGY | Office of Science

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.

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 were renewals of existing EFRCs and 10 of which were new EFRCs. In 2016, DOE added 4 new four-year centers to accelerate the scientific breakthroughs needed to support the Department's environmental management and nuclear cleanup mission, bringing the total number of active EFRCs to 36. An open recompetition in 2018 resulted in 42 awards: 11 two-year extensions of existing EFRCs, 9 four-year renewals of existing EFRCs, and 22 four-year awards for new EFRCs. The technical research summaries in this document describe the planned work, partner institutions and associated senior investigators of the 46 EFRCs that are active in 2018 – 2020. The index at the end of the document includes keywords, grand challenges, transformative opportunities, and references to BES reports that the leadership of each EFRC has identified as relevant to their centers.

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Ordered first by Institution and then by Director

Center for the Advancement of Topological Semimetals (CATS)
EFRC Director: Robert McQueeney
Lead Institution: Ames Laboratory
Class: 2018 – 2022

Mission Statement*: To understand and discover new quantum phenomena and functionality in topological materials for future applications in spin-based electronics, computing, and sensing.*

Recent theoretical predictions and experimental discoveries of topological semimetals (TSMs), an entirely new class of materials, are opening an exciting frontier of science at the intersection of magnetism and topology. TSMs are poised to trigger breakthroughs in dissipationless spin and charge transport, such as high temperature quantum spin Hall and quantum anomalous Hall effects. Special TSMs, called Weyl semimetals (WSMs), can host extreme magnetotransport and optical properties, optically and magnetically switchable states, and gate-tunable chiral plasmonics. Understanding the basic principles of TSMs may also deliver new materials platforms for mid-infrared photodetection, night vision, and light harvesting. Research in this incipient field is fueling enthusiasm that TSMs will ignite transformational opportunities in spintronics, optoelectronics, quantum sensing, and classical (Beyond Moore's Law) and quantum computing.

To realize this potential, we need to discover and understand the properties of archetypal *magnetic* TSMs, including magnetic WSMs, magnetically proximitized Dirac semimetals (DSMs), and two-dimensional (2D) spin-polarized DSMs. Magnetic TSMs provide the exciting potential of harnessing magnetic fields and magnetic interactions to control the flow of both charge and spin and their interconversion. We need to develop methods to assemble atomically thin 2D layers and thin films of TSM into heterostructures with other magnetic materials to deliver functionality and even induce new topological states of matter. The basic principles for the control and manipulation of TSM functionalities need to be unveiled by studying the interaction of TSM materials with external fields.

With leading expertise in the theory of TSM, new materials discovery, the assembly of heterostructrures, and advanced characterization methods, CATS advances our understanding and accelerates breakthrough innovations in topological semimetals. CATS pursues **three integrated, fundamental research goals** that address essential issues in TSM research:

1. *Predict, discover, and understand archetypal magnetic TSMs and new magnetic topological states-of-matter.* There is a need to discover new magnetic TSM compounds with simpler topology and the absence of trivial bands near the Fermi energy. Also, the combination of TSM with other materials in heterostructures can deliver new quantum topological states. CATS addresses these materials challenges with a systematic approach described below.
2. *Controllably induce topological phase transitions.* An important milestone in CATS is a clear demonstration that topological properties can be controlled. This requires the ability to create, modify, or annihilate the topologically protected electronic states. CATS uses a multitude of methods to enable TSM control, such as carrier doping, electrostatic gating, magnetic fields, strain and dimensionality (in thin films), and proximity effects (in heterostructures).
3. *Manipulate the response of topological states to external fields.* Demonstrating that TSM states can be manipulated is the first step toward fulfilling their promise in information and sensing technology. One route to achieve this goal is to take advantage of gapless topological bands and utilize their unique light-matter and optoelectronic responses to manipulate and switch

charge/spin currents or to generate collective electronic effects. CATS researchers exploit the unique pulsed high magnetic fields and ultrafast coherent photon excitations to manipulate TSMs and potentially discover new states of matter.

To attain these goals, CATS employs innovative approaches that are organized into **three crosscutting research thrusts (RTs) to**: **(1) predict, discover, and understand new bulk magnetic TSMs; (2) discover and control novel quantum states and functionality in thin films and heterostructures; and (3) investigate the dynamical manipulation of topological states.** The goals and RTs of CATS are interwoven. For example, the discovery of new topological states of matter (Goal 1) may be discovered in bulk single crystals (RT-1) or heterostructure assemblies (RT-2), or under non-equilibrium conditions in applied fields (RT-3). In addition, the development of emergent functionality in layered heterostructures of 2D materials (RT-2) requires the discovery of new, exfoliatable TSMs (RT-1).

Developing the unique functionalities of TSMs is a *complex materials challenge.* CATS will discover archetypal TSM compounds and deliver high-quality, well-characterized materials to be used as building blocks in heterostructures. All research thrusts in CATS exploit premier synthesis and characterization capabilities and the ability to apply static and time-dependent external fields to manipulate and switch TSM properties. CATS combines fundamental theory and insight with first-principles electronic structure calculations; results from such calculations for relevant materials and heterostructures will be used to construct response functions, magnetotransport models, and non-equilibrium theories. CATS also utilizes DOE-supported neutron sources, light sources, nanocenters, and leadership computing user facilities.

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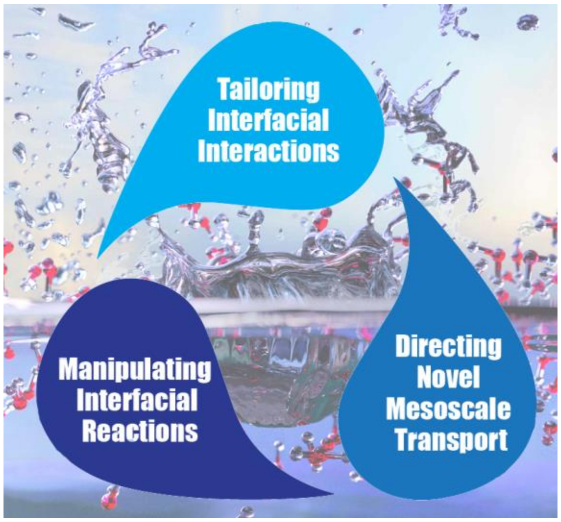
Advanced Materials for Energy-Water Systems (AMEWS)
EFRC Director: Seth Darling
Lead Institution: Argonne National Laboratory
Class: 2018 – 2022

Mission Statement*: To understand and design water-solid interfaces to enable future advances in materials for efficient water treatment.*

The direct connection between water and energy takes its most tangible scientific form at water/solid interfaces that mediate energy conversion and transduction processes, or are designed to influence water chemistry. Water/solid interfaces are central to a broad array of scientific and technological processes including heterogeneous catalysis and electrochemistry, life sciences and biomedical applications, and environmental and geosciences. The importance of such systems cannot be overstated. Yet, numerous fundamental questions in these areas remain unanswered despite decades of study. At the heart of these issues are molecular-scale questions involving the nuances of water's hydrogen bonding at interfaces with electrolyte solutions, the interfacial transfer of energy in the form of protons and electrons, the adsorption and chemical reactivity of solutes at structured and confined interfaces, and many other particulars connected to water. With the emergence of newfound capabilities to experimentally probe and computationally model these deceptively complex systems, the chemistry and physics of aqueous solution/solid interfaces has become one of the most exciting fields in science.



The AMEWS team brings together a confluence of capabilities to tackle the knowledge gaps outlined above. We have identified three integrated 4-year goals toward which we will work collectively as a center:

- Design and synthesize responsive interfaces to selectively and reversibly adsorb specific components from a complex aqueous fluid.
- Decipher and harness the interplay between confinement and charge on catalytic reactivity at water/solid interfaces.
- Predictively describe the transport of water, aqueous solutions, and charged species across multiple time and length scales, especially under extreme confinement and in the presence of charged interfaces.

These goals target the three legs of water/solid interfaces: adsorption, reactivity, and transport. Our first goal aims to understand interactions between constituents of a multi-component aqueous fluid and a solid interface sufficiently well to enable selective adsorption of particular species in the solution while rejecting others. Further, we aim to design these affinity and repulsive interactions with precision such that they can be switched or tuned for reversible adsorption. We will explore how electrostatics, hydrogen bonding, surface chemistry, and microstructure influence interfacial affinity and the organization of the solution's boundary layers.

Confinement and charge have long been investigated in the context of reactivity and catalysis in aqueous systems, but emerging computational and experimental capabilities enable unprecedented potential for progress in understanding these essential phenomena. AMEWS will explore the explicit role of water in reactive processes on the surfaces and near-surface of solid/liquid interfaces, particularly (electro)catalytic processes. We will study the effect that precise spatial confinement (including electric fields) may have on catalytic function in complex aqueous media, relying on unprecedented control of hierarchical architectures over length scales from Ångstroms to microns and using advanced in situ and time-resolved spectroscopic methods. We will also utilize our ability to design and synthesize atom-precise catalysts to investigate the reactions in complex aqueous media on designer reactive interfaces.

Macroscopic aspects of aqueous solution transport are reasonably well understood, but as the dimensions of the channel through which the fluid flows approach the molecular scale and interface effects increase in prominence relative to the bulk, current models fail to predictively capture transport behavior. This is particularly true when the interfaces are charged and interact electrostatically with ions and polar (or polarizable) species in the solution—including water itself. Building predictive models will only be possible if the various interactions are well understood at each of the relevant length scales at play. Experimentally, AMEWS will work toward this goal by applying new methodologies such as directed self-assembly—coupled with our expertise in atomic layer deposition—to generate extremely well-defined nanoporous media in which the pore diameters are nearly monodisperse and the pore wall chemistry and charge can be tuned at will.

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Center for Electrochemical Energy Science (CEES)

EFRC Director: Paul Fenter
Lead Institution: Argonne National Laboratory
Class: 2009 – 2020

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.*

The Center for Electrochemical Energy Science (CEES) EFRC seeks to enable significant advances in energy storage technology through a concerted and broad-based science research program. This program is designed to create a foundational understanding of the structures and electrochemical processes that limit the technological properties and advantages of lithium ion battery (LIB) systems with a central focus on the behavior of metal oxide cathode materials used in today’s LIB cathode systems. The CEES program addresses two important challenges that confront LIBs: 1) LIB lifetime and safety are strongly influenced by secondary reactions that occur primarily at the electrode-electrolyte interface. The inability to completely control electrochemical reactivity and associated side reactions is reflected by capacity loss through electrolyte decomposition, active materials dissolution and structural decay, and oxygen evolution with the possibility of fire and/or explosion. 2) The energy density of current LIB systems is limited predominantly by the inherent electrochemical capacity of lithium-intercalated metal oxide cathodes. Various “beyond Li-ion insertion” chemistries can, in principle, exceed the cathode capacity and energy density of current LIBs, but these reactions generally involve phase transitions that are kinetically challenging and suffer from poor reversibility and significant over-potentials. Within this context, CEES research efforts are designed to: develop a robust understanding of the electrochemical reactivity of metal oxide cathode materials relevant to lithium ion battery technology; leverage this understanding to control the associated lithiation/delithiation processes; and, explore new conceptual approaches and materials that may be used to advance present-day LIB systems.

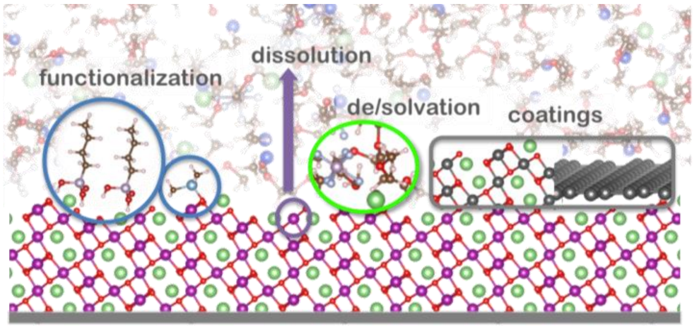

Figure 1: Interfacial coatings and modifications to control the reactivity of LMO-electrolyte interfaces.

CEES focuses on the structure and reactivity of the electrode-electrolyte interface. One set of objectives is to understand the molecular-scale structure and reactivity of simple oxide-electrolyte interfaces, including both primary reactivity of reversible lithiation, and secondary reactivity associated with capacity loss and electrode decomposition. This activity will develop a mechanistic understanding of how interfacial coatings and surface modifications alter the reactivity of $LixMn_2O_4$ (“LMO”). This work will build on the foundational understanding of the reactivity of model LMO(111) thin-film cathodes that we have explored both computationally and experimentally, through the use of model systems and state of the art experimental and computational approaches with an emphasis on developing conceptual advances. Parallel to this effort will be studies to understand the elementary steps in the modification of LMO electrode surfaces with atomic layer deposition, such as in ongoing work with alumina coatings. These studies will leverage the CEES capabilities to directly and independently probe both stress and strain evolution in LMO electrodes enabling new insights into the thermodynamic and kinetic controls over these processes and the development of a new concept, “electrochemical stiffness” with ongoing work exploring how coatings influence stress/strain development. These insights compliment ongoing studies of model LMO thin-film reactivity in which preliminary results suggest that strain modifies the lithiation pathway for laterally constrained (i.e., epitaxial) LMO thin films.

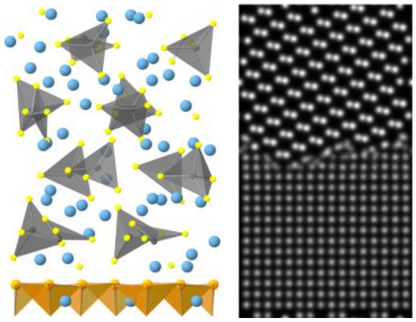

Figure 2: A model interface between a solid-electrolyte and a solid electrode, and a simulated transmission electron microscopy image.

A second theme seeks to define and implement strategies for robust interfacial modification that can control secondary reactivity at elevated potentials. This effort focuses on the development of structure-function relationships with respect to the surface modification of LMO interfaces to control its reactivity. We will extend these capabilities to stabilize the cathode-solid electrolyte interface. This work builds on earlier CEES work on the active functionalization of model LMO surfaces and related interfaces. Our hypothesis in these studies is that the interface separating solid-electrolytes from an electrode is a key weak-link whose structure and reactivity is largely unknown. It is postulated that: 1) the atomistic interfacial structure controls the rate limiting steps for lithium ion transfer; 2) evolution of these interfaces is due to intrinsic chemical/electrochemical instabilities associated with chemical and physical disequilibria; and 3) interface stabilization requires the development of new materials and design strategies that provide stability while retaining functionality. Based on these postulates we will identify the atomistic structures and instabilities at the interfaces through the study of selected solid electrolytes with LMO and their relationship to mechanisms that control ion transfer. These insights will be used to guide the creation of interlayers that achieve stability and functionality at solid electrolyte interfaces in the presence of disequilibria.

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Northeast Center for Chemical Energy Storage (NECCES)
EFRC Director: M. Stanley Whittingham
Lead Institution: Binghamton University
Class: 2009 - 2020

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 scientific research goals are to:

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 and sodium.
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, including the understanding of the role of anion redox in intercalation cathodes.

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:

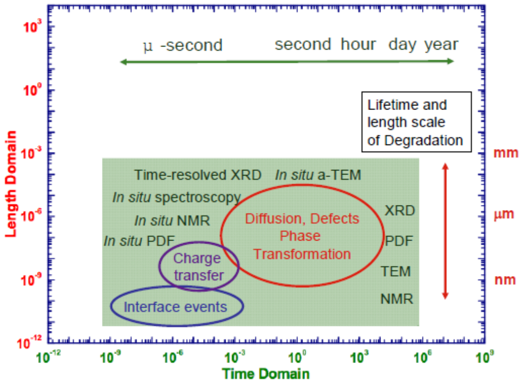
(i) The structural parameters required for rapid ionic (Li^+) motion (and thus high rate), particularly at close to full lithium removal.
(ii) The minimal electronic (and ionic conductivity), required to completely extract Li from materials for different crystal structures and particle sizes.
(iii) How to control (minimize) structural changes so as to enable redox processes involving more than one electron.
(iv) What determines the contribution of anion redox in intercalation reactions, and how it could be optimized.
(v) 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 involves the development of novel *operando* and *ex-situ* experimental approaches aimed at probing electrical energy storage materials at the atom, single crystal/particle, and across the electrode heterostructure. 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. 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, operando TEM, high energy resolution STEM-EELS.

(iii) *Imaging reactions across the electrodes hetero-structures*: 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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Molten Salts in Extreme Environments (MSEE)
EFRC Director: James Wishart
Lead Institution: Brookhaven National Laboratory
Class: 2018 – 2022

Mission Statement*: To provide a fundamental understanding of molten salt bulk and interfacial chemistry that will underpin molten salt reactor technology.*

Molten Salt Reactors (MSRs) are a potentially game-changing technology that could enable cost-competitive, safe, and more sustainable commercial nuclear power generation. Proposed designs employ molten salts in the temperature range of 500 – 900 ˚C acting as coolants for solid-fueled reactors or in other cases where the nuclear fuel dissolved in the molten salt as combined coolant and fuel. Consequently, the development of reliable MSRs requires a comprehensive understanding of the physical properties and chemistry of molten salts and of their interfacial interactions with reactor materials.

The Energy Frontier Research Center for Molten Salts in Extreme Environments (MSEE) will provide fundamental and predictive understanding of the bulk and interfacial chemistry of molten salts in the operating environments expected for MSRs. MSEE addresses this challenge through a coordinated experimental and theoretical effort to elucidate the atomic and molecular basis of molten salt behavior, including interactions with solutes (dissolved materials such as nuclear fuel and fission products) and interfaces, under the coupled extremes of temperature and radiation.

The research of MSEE is organized into two interrelated thrusts. The first is *Molten Salt Properties and Reactivity*, which aims to understand how molecular-scale interactions, structure and dynamics lead to macroscale properties. A key focus is to learn how the interactions between molten salts and solutes affect physical properties and control solubility and reactivity. The second thrust, *Interfacial and Corrosion Processes in Molten Salt Environments*, aims to understand the atomic-scale structure and dynamics at interfaces and related mechanisms of interfacial and corrosion processes between molten salts and materials, including the effects of extreme environments such as radiation and high temperature.

Thrust 1: Molten Salt Properties and Reactivity

Aim 1: Determine the structure and dynamics of molten salt solutions across scales of length and temperature. Powerful X-ray, neutron-scattering and optical spectroscopy techniques are employed and coupled with computational approaches to interpret observations and validate predictions in order to assemble a dynamic model of molten salt structure.

Aim 2: Elucidate the principles that control metal ion solvation, speciation and solubility in molten salts. The same methods are used to understand changes in solution structure, dynamics and thermal properties when solutes, including actinides and fission products, are dissolved in molten salts.

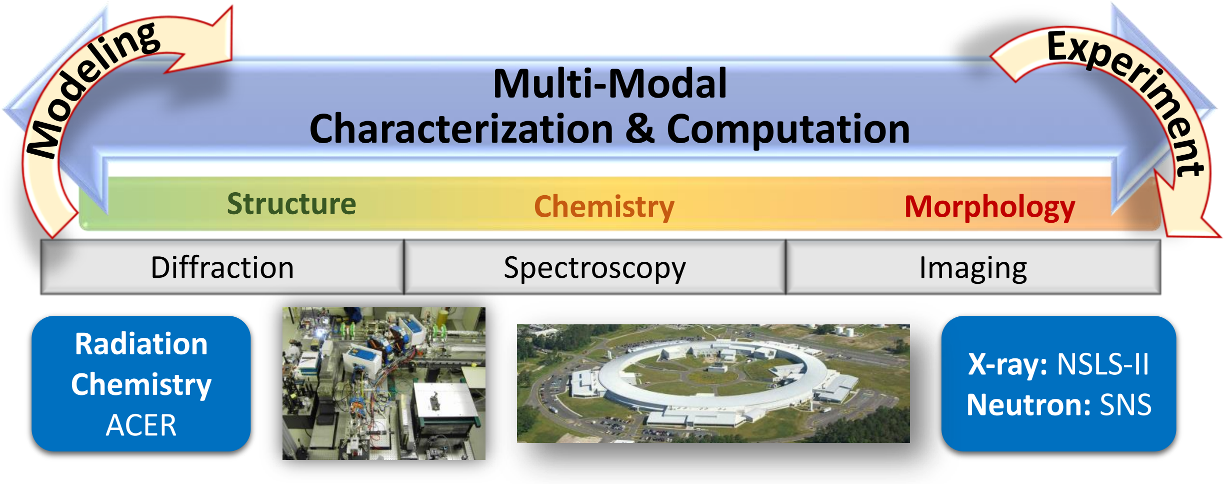
Aim 3: Understand how radiation affects salt solution chemistry and solute speciation. Radiation chemistry techniques are used to examine the radiation-driven reactions of molten salts and materials dissolved in them.

Thrust 2: Interfacial and Corrosion Processes in Molten Salt Environments

Aim 1: Measure and predict the structures and dynamics of molten salts at interfaces. X-ray and neutron reflectivity measurements are integrated with new modeling approaches to provide fundamental new information on surface ordering and dynamics of molten salts and to elucidate how these structures determine energy and charge transfer across the interface.

<u>Aim 2: Kinetics of interfacial reactions leading to corrosion.</u> In-situ experimental techniques, enabled by advances in characterization capabilities, will provide unprecedented temporal and spatial resolution for quantifying interfacial reactions and help us understand and predict non-equilibrium, metastable states formed during the reactions at interfaces.

A deeper knowledge on molten salt structure and properties, and the behavior of the actinides, fission products and corrosion products in molten salt solution under radiolytic conditions, will strengthen the scientific foundation for the practical implementation of MSRs. A stronger understanding of redox chemistry and solvation of solutes such as fuel metal ions and fission products will contribute to better predictions of precipitation, participation in corrosion reactions, gas generation and failure to behave as desired in fission product separations. Improved molecular knowledge of the corrosive interactions of molten salts will suggest ways to mitigate challenges to the performance of nuclear reactor materials, and also in solar thermal collectors. MSEE will focus on filling those knowledge gaps to enable safer, higher performing and more reliable MSR systems, as well as to extend our scientific understanding of the general fundamental chemical processes in molten salts.



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Breakthrough Electrolytes for Energy Storage (BEES)
EFRC Director: Robert Savinell
Lead Institution: Case Western Reserve University
Class: 2018 – 2022

Mission Statement*: To develop fundamental understanding of: (i) solvation and transport properties; (ii) electrode-electrolyte interfaces; and (iii) electron transfer reactions in deep eutectic solvents and soft nanoparticle electrolytes.*

Discovery of new electrolytes is needed for advancing the fundamental science and enabling new opportunities in electrochemical systems including redox flow batteries, supercapacitors, electrocatalysis, electrodeposition, separations and sensors. Specifically, by designing new electrolytes with higher concentrations of electrochemically active species, lack of flammability and ease of control over transport properties, substantial improvements will be realized in energy and power density, safety and reductions in environmental impact, and cost of energy storage systems. The Breakthrough Electrolytes for Energy Storage (BEES) EFRC sets out a comprehensive research program (Figure 1) that leverages expertise in the theory-guided synthesis of novel materials, and the characterization of their properties using simulations and experiments. By understanding the role of chemical structure on physicochemical properties of new electrolytes, efficient electron and charge transfer processes will be enabled.

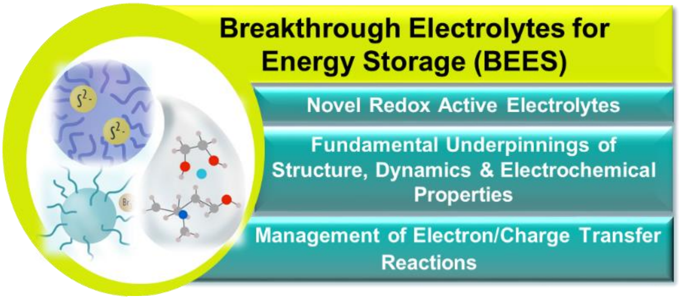

Figure 1. Research overview of BEES

The two research thrusts within BEES are: (1) Deep Eutectic Solvents (DES) and (2) Soft Nanoparticles (SNP):

Thrust 1: **DES** are a class of liquids comprised generally of a hydrogen bond donor, like a halide salt, and a hydrogen bond acceptor. DES are non-toxic, biodegradable, stable, nonvolatile, and nonflammable. They have a high degree of structural flexibility. DES enable electrochemical reactions without the constraints of aqueous solvents. <u>The goal of Thrust 1</u> is to unravel the fundamental underpinnings of the relationship between the composition and structure that determine the physicochemical and electrochemical properties of DES. With this understanding functionalized DES with redox active groups will be created as new electrolyte systems, improving redox-active material solubility and facilitating fast interfacial electron transfer reaction rates.

The main <u>hypothesis of Thrust 1</u> is that spatial and dynamic heterogeneity introduced by noncovalent interactions alters the molecular energy landscape and leads to mesoscale organization and dynamics that determine the macroscopic properties of DES. Within the scope of BEES, we will answer the following **scientific questions**:

1. *Is the extent of hydrogen bonding (number, strength, lifetime) in DES a surrogate measure of diffusivity and conductivity of ions?*
2. *Is there a correlation between the melting point, molar free volume and viscosity of DES?*
3. *How are DES structured near the interface? What kind of screening lengths should we expect?*
4. *What are the impacts of surface adsorbed species and metal speciation on the kinetics and reversibility of electron transfer reactions between an electrode and a redox-active DES?*
5. *What are the key structural features of DES that control its reactivity near charged surfaces?*

The answers to these questions will be the basis for (i) tailoring DES structures for specific electrochemical and transport properties, and (ii) extending the electrochemical stability of the DES structures over wide potential windows to enable new electrochemical reactions not feasible in traditional DES systems.

Thrust 2: **SNP** electrolytes are heterogeneous, multiphase systems where liquid droplets are dispersed in a carrier phase. An example of a SNP electrolyte is nano-emulsion in which droplets containing electroactive species are surrounded by a fluid that provides conductivity. Another example is a NOHM (Nanoparticle Organic Hybrid Materials) which are liquids formed from hard nanoparticles with attached, possibly functionalized, polymeric chains. The goal of Thrust 2 is to enable unique paths for decoupling the nature and solubility of electroactive material from the conductivity and transport of ions in the surrounding solution, which may be an aqueous phase or a non-aqueous phase.

The guiding hypotheses for Thrust 2 are (i) that microemulsions and NOHMs can controllably take up and release redox active species and (ii) that SNP-contained redox active species can be converted through direct or mediated electron transfer across the boundaries of the SNP. The following **scientific questions** will be answered:

6. *How does the introduction of an ionic backbone or polar functionalities into polymers or surfactants used in NOHM and microemulsions affect the mobile ion packing and dynamics of these systems?*
7. *How does the introduction of small ions into the NOHM and microemulsions affect the packing and dynamics of polymers or surfactants?*
8. *How does their immersion in electrolyte solutions affect the transport behavior of SNP?*
9. *How do electron and ion transfer rates into microemulsions and the canopy of NOHM depend on the structure and dynamics of polymers or surfactants?*
10. *What interactions drive uptake of solutes into SNP?*

The answers to these questions will be the basis for tailoring new structures for specific electrochemical and transport properties as well as for enabling uptake of electroactive species. The overarching aim will be to develop the electrochemical science underpinnings of SNP electrolytes.

Breakthrough Electrolytes for Energy Storage (BEES)	
Case Western Reserve University	Robert Savinell (*Director and Research Integration Officer*), Rohan Akolkar (*Deputy Director*), Clemens Burda, Burcu Gurkan (*Thrust 1 Lead*), Jesse Wainright
Brookhaven National Laboratory	Radoslav Adzic, Miomir Vukmirovic
Columbia University	Ah-Hyung (Alissa) Park
Hunter College	Steven Greenbaum
New York University	Mark Tuckerman
Notre Dame University	Edward Maginn (*Deputy Thrust 1 Lead*)
Texas A&M University	Emily Pentzer
University of Tennessee, Knoxville	Tessa Calhoun, Mark Dadmun, Douglas Hayes (*Deputy Thrust 2 Lead*), Joshua Sangoro, Thomas Zawodzinski (*Thrust 2 Lead*)
University of Texas at Austin	Allen Bard

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Programmable Quantum Materials (Pro-QM)
EFRC Director: Dmitri N. Basov
Lead Institution: Columbia University
Class: 2018 – 2022

Mission Statement*: To discover, characterize, and deploy new forms of quantum matter controllable by gating, magnetic proximity and nano-mechanical manipulation.*

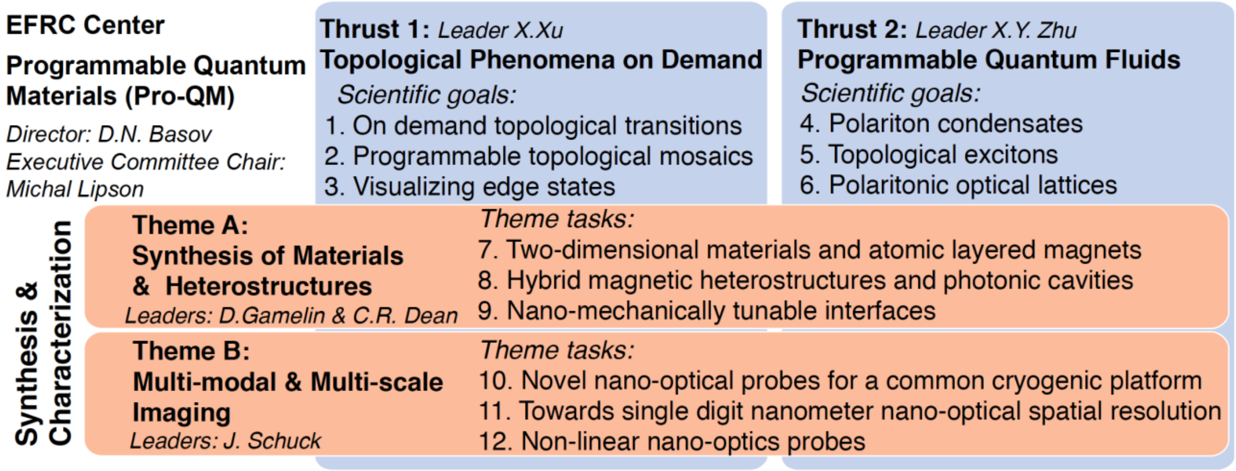
Experimentally realizing quantum phases of matter and controlling their properties is a central goal of the physical sciences. In this endeavor, fundamental science is particularly relevant to technological advances. Novel quantum phases with controllable properties are essential for new electronic, photonic, and energy management technologies needed to address the growing societal demands for rapid and energy efficient information processing and transduction. *Quantum materials (QMs*) offer particularly appealing opportunities for the implementation of on-demand quantum phases. QMs host interacting many-body electronic systems featuring an intricate interplay of topology, reduced dimensionality, and strong correlations that leads to the emergence of "quantum matter'' exhibiting macroscopically observable quantum effects over a vast range of length and energy scales.

The unified four-year scientific goals of Pro-QM are organized in two interdependent Research Thrusts (Fig. 1). Thrust-1 will *create, visualize, and utilize intertwined, controllable, and interacting topologically protected states in quantum materials.* Thrust-2 will *create, manipulate, and understand macroscopic coherent states and induce transitions to novel quantum phases.* Both Thrusts explore optically driven effects where light creates new states not present at equilibrium (transient edge states in Thrust 1 and excitonic states in Thrust 2) and examine complementary topological phenomena (mainly electronic in Thrust 1, and mainly excitonic in Thrust 2). The Thrusts harness and rely on two major research Themes, built on the cross-cutting strengths of our team: (A) the creation of new tailored materials and architectures to understand and exploit interfaces, and (B) transformative advances in experimental imaging tools for probing optoelectronic and magnetic properties at their native length- and time-scales. The concerted EFRC effort is therefore imperative to make the desired leaps in progress.

Programmable QMs properties are essential for realizing the promise of quantum technology for disruptive advances in information transfer, processing, sensing, and other currently unimagined functions.

Our team will focus on transition metal dichalcogenides (TMDCs) and 2D-halides: two representative classes of layered van der Waals (vdW) solids combining novel properties with an unprecedented degree of controllability. Realizing the potential for programmable quantum matter requires a three-pronged approach, combining *i)* the unique suite of controls and driving perturbations, with *ii)* a transformative set of synthesis/device fabrication capabilities (Theme A) and *iii)* new nanoscale characterization techniques integrated in a single platform (Theme B). These strategies are particularly well-adapted to vdW materials. Our approach is to combine the three prongs into one cohesive team effort, expanding on already strong collaborations within the Pro-QM team.

Our chief scientific goals and tasks are outlined in Fig. 1 and closely aligned with the DOE Grand Challenges and Basic Research Needs Reports. A common thread underpinning these clear but ambitious goals and tasks is to develop strategies for transforming QMs into a desired state with tailored quantum properties not attainable in common metals or semiconductors. The present knowledge gaps remain immense but can be effectively addressed given the unique combined expertise of the Pro-QM team documented through a track record of breakthrough collaborative research.



EFRC Center

Programmable Quantum Materials (Pro-QM)

Director: D.N. Basov
Executive Committee Chair: Michal Lipson

Thrust 1: *Leader X.Xu*
Topological Phenomena on Demand
Scientific goals:
1. On demand topological transitions
2. Programmable topological mosaics
3. Visualizing edge states

Thrust 2: *Leader X.Y. Zhu*
Programmable Quantum Fluids
Scientific goals:
4. Polariton condensates
5. Topological excitons
6. Polaritonic optical lattices

Synthesis & Characterization

Theme A:
Synthesis of Materials & Heterostructures
Leaders: D.Gamelin & C.R. Dean
Theme tasks:
7. Two-dimensional materials and atomic layered magnets
8. Hybrid magnetic heterostructures and photonic cavities
9. Nano-mechanically tunable interfaces

Theme B:
Multi-modal & Multi-scale Imaging
Leaders: J. Schuck
Theme tasks:
10. Novel nano-optical probes for a common cryogenic platform
11. Towards single digit nanometer nano-optical spatial resolution
12. Non-linear nano-optics probes

Figure 1: Energy Frontiers Research Center on Programmable Quantum Materials. Center activities are organized into two Thrusts and two cross-cutting Themes.

Programmable Quantum Materials (Pro-QM)	
Columbia University	Dmitri N. Basov (Director), Ana Asenjo Garcia, Cory R. Dean, James Hone, Michal Lipson, Andrew J. Millis, Abhay Pasupathy, P. James Shuck, Xiaoyang Zhu
University of Washington	Jiun-Haw Chu, David H. Cobden, Daniel Gamelin, Xiaodong Xu
Carnegie Mellon University	Di Xiao

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Center for Alkaline Based Energy Solutions (CABES)
EFRC Director: Héctor D. Abruña
Lead Institution: Cornell University
Class: 2018 – 2022

Mission Statement*: To achieve a detailed understanding of the nature, structure, and dynamics of electrocatalysis in alkaline media.*

CABES will integrate theory and computational methods for catalysis and interfacial structure/dynamics; the synthesis of model (electro)catalytic systems with atom-level control; ionically conducting/transporting polymers/membranes; catalyst support systems and architectures in contact with metal electrodes; computational materials science to guide the synthesis of next-generation materials; and the development of experimental tools that will provide *in situ/operando*, spatiotemporal characterization of systems under operation. The proposed studies are aimed at significantly advancing electrocatalysis in alkaline media through the rational design and development of new materials and architectures as well as experimental and computational tools necessary for, and critical to, a fundamental understanding of these processes. Results will, in turn, impact numerous technologies, including alkaline fuel cells, electrolyzers, and all metal/air batteries.

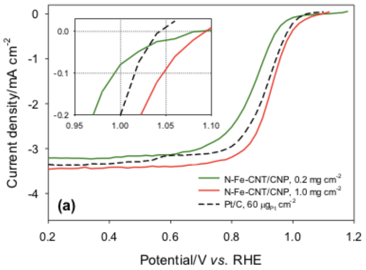

Fig. 1: ORR performance of N-Fe-CNT/CNP composite and Pt/C catalysts. Main Panel: RDE polarization plots in 0.1 M NaOH at 25°C and at 900 rpm. Inset: expanded view of the low overpotential region.

CABES will focus on: (1) <u>Electrocatalysts</u>: We will develop ORR electrocatalysts that exhibit high activity and long life in *alkaline* media with initial focus on ordered intermetallic structured cores and shape-controlled nanoparticles. We will explore the electrocatalytic activity of transition metal oxides and nitrides as well as PGM-free ORR electrocatalysts (Fig. 1). For the HOR, we will use, as a point of departure, recent findings on the HOR activity of IrRu/C, IrPd/C, and IrPdRu/C alloy nanoparticle catalysts. (Fig. 2).

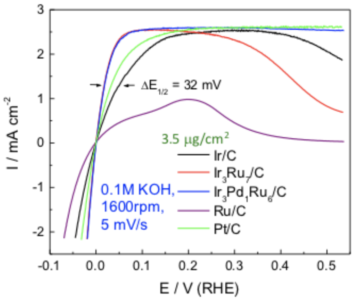

Fig. 2: RDE voltammograms of IrRu/C and IrPdRu/C in H_2 saturated 0.1 M KOH. Scan rate: 5 mV/s, rotation rate: 1600 rpm. The catalyst loading was 3.5 $\mu g_{metal}/cm^2$. Alloy catalyst compositions are indicated.

(2) <u>Support systems:</u> We will develop strategies for the design, synthesis and characterization of catalyst supports that are conductive and stable at high potentials under alkaline conditions. We will focus on the design, synthesis and characterization of hierarchical porous carbons as model catalyst supports in alkaline media. We will evaluate new hierarchical motifs to harness their complex functionality. We will take advantage of recent findings that Nb-doped rutile TiO_2 supports retain their electronic conductivity, even under the strongly oxidizing electrochemical conditions of a fuel cell or electrolyzer. We will also investigate mesoporous transition metal nitrides and oxy-nitrides. We will pursue a strategy for modeling the impedance of a supported catalyst by a transmission line model, using Kirchhoff's law current loops to model the serial-parallel flow of current (Fig. 3).

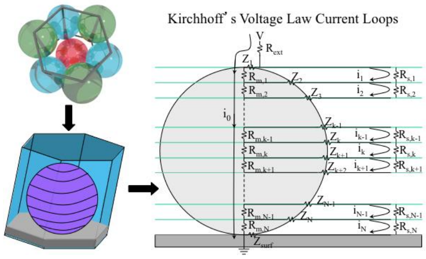

Fig. 3: Unit cell model of oxide support particles in a catalyst/ support/ electrolyte film, and Kirchhoff's law decomposition of the components of the film's impedance.

(3) Alkaline membranes: CABES will develop methods to synthesize phosphonium- and imidazolium functionalized norbornenes and *trans*-cyclooctenes. These monomers are unique because the ring strain of norbornenes and *trans*-cyclooctenes is higher than *cis*-cyclooctenes and consequently, these functionalized building blocks can be polymerized in a living fashion. We will also seek to understand the mechanism of membrane degradation and potential carbonate formation and precipitation, as they will guide the structural design and optimization of ionomers with improved stability and high intrinsic conductivity.

(5) Theory: Our theory and computational efforts will involve the elucidation of intricate chemical-reaction and electron-transfer (ET) pathways as well as fundamental understanding of complex electrochemical environments (Fig. 4) that include externally applied voltages, solvent, mobile ions, the role of interfacial water and ions, mechanistic pathways such as proton-coupled electron transfer (PCET), and reactive intermediates.

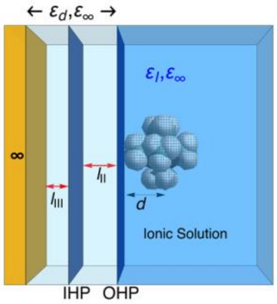

Fig.4 Dielectric continuum description of an electro-chemical interface. CABES will use first-principles simulations of the electrode-liquid interface to extract the information necessary to compute reorganization energies for complex systems.

(6) Analytical Methods: CABES will take advantage of its extensive expertise in the development and use of *in situ* and *operando* methods for establishing a synthesis/characterization feedback loop in engineering heterogeneity i.e., interfaces, defects, strains, confinement, charge-carrier distribution, and concentration of intermediate species for electrocatalytic reactions. We will carry out studies on multiple length scales from atomic-resolution maps of composition and bonding in catalyst nanoparticles, 1-2 nm resolution *operando* STEM and EELS, nanoscale 3-D microscopy with small (micro) X-ray beams (Fig. 5). We will employ a new EMPAD detector to map out strain on nanoparticle catalysts. The use of cryo-TEM will enable imaging of radiation-sensitive membranes.

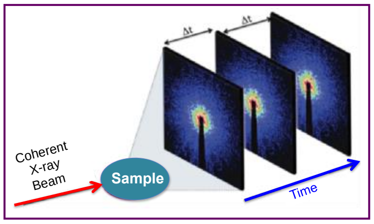

Fig. 5 Schemtaic of X-ray Photon Correlation Spectroscopy to study dynamics of catalytic surfaces.

Summary: Over the four-year period, we foresee the CABES effort as providing the basis for ushering in an Alkaline-Based Energy Technology Society.

Center for Alkaline Based Energy Solutions (CABES)	
Cornell University	Héctor Abruña (Director), Tomas Arias (Thrust Area Leader), Geoffrey Coates (Thrust Area Leader), David Muller (Thrust Area Leader), Joel Brock, Peng Chen, Francis DiSalvo, Robert DiStasio, Emmanuel Giannelis, Lena Kourkoutis, Andrej Singer, Jin Suntivich, Paul Mutolo (Executive Director)
University of Pennsylvania	Tom Mallouk (Thrust Area Leader)
University of Binghamton	Jiye Fang
University of Wisconsin	Manos Mavrikakis
Carnegie Melon University	Kevin Noonan
Los Alamos National Lab	Piotr Zelenay
National Renewable Energy Lab	Bryan Pivovar
Yale University	Sharon Hammes-Schiffer

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Center for Actinide Science & Technology (CAST)
EFRC Director: Thomas E. Albrecht-Schmitt
Lead Institution: Florida State University
Class: 2016 – 2020

Mission Statement*: To advance our understanding of how electronic structure and bonding control the properties of radioactive materials. This knowledge will aid in the development of nuclear technologies that enhance energy security, address nuclear legacy issues and environmental concerns, and foster the next generation of nuclear scientists.*

Cultivating new technologies that address the challenges associated with nuclear waste generated during the Cold War requires an advanced, science-based understanding of *f*-element (lanthanide and actinide) behavior both in solution and in the solid state. The Center for Actinide Science & Technology (CAST) brings together scientists with *f*-element expertise from Florida State University, the National High Magnetic Field Laboratory, Florida International University, Los Alamos National Laboratory, Lawrence Berkeley National Laboratory, Purdue University, the University of Pennsylvania, the University of Manitoba, and SUNY Buffalo to create synergy and cross-fertilization between chemists, physicists, materials scientists, and theoreticians. We are developing unique expertise in synthesis, characterization, and theory in order to create new materials that provide unprecedented chemical selectivity for specific radionuclides. An important aspect of this research is developing experimentally-verified theoretical methods for understanding complex chemistry and physics. These advancements in experiment and theory will expand our knowledge of actinide materials under normal and extreme conditions. In addition to the practical applications of this research, an improved understanding of these enigmatic elements will expand the boundaries of the periodic table.

Synthesis of Solid-State and Molecular Materials. The actinide materials and molecular systems that are the focus of CAST are being prepared using the unique synthetic capabilities at FSU, LANL, and LBNL. A host of physical property measurements on these materials are taking place at the NHMFL. This allows us to probe properties under a variety of extreme conditions including high magnetic fields (the highest in the world), temperatures, and pressures. Similarly, the synthesis of novel actinide materials and molecules is being lead by FSU, LANL, LBNL, and other partner institutions. The preparative efforts include the design, synthesis, and optimization of new and existing ligands needed for new transuranium separations. The ligand design and complexation studies with lanthanides, thorium, and uranium primarily occurs at the University of Pennsylvania and Purdue University, and is then transferred to institutions with transuranium laboratories. The capabilities of FSU and LANL to work with macroscopic quantities of transuranium elements, such as plutonium, for basic research purposes are almost unique. As such, CAST is focused on directly probing materials of interest such as plutonium solids and americium coordination complexes as opposed to using surrogates for these elements that often provide an incomplete or sometimes even inaccurate predictions for real radioactive molecules and materials. These materials are fully characterized using all available structural and spectroscopic methods that one would normally employ to understand non-radioactive materials. Thus, some of CAST's experimental efforts are devoted to developing safe methods for performing these measurements at user facilities. Obtaining the highest quality experimental data is essential not just to our experimental understanding, but also provides critical data for benchmarking new theoretical methods.

Electronic Structure Theory. There are no broadly available, reliable, and predictive electronic structure approaches for strongly-correlated materials. We are expanding our approaches to employ high-accuracy electronic structure methods for predicting the properties of actinide compounds. Results from these calculations are validated by comparison with experimental results including those from X-ray absorption spectroscopy and magnetic susceptibility measurements. Of particular importance is the fact that actinide elements possess electrons that are travelling at significant fractions of the speed of light. This causes their mass to increase, the shape of orbitals to change, and reorganization of the orbital energies. Historical computational methods have not been up to the task of tackling the large number of electrons and the large number of orbitals involved in bonding in actinide coordination complexes or materials. Moreover, the importance of relativistic effects in these systems has also been neglected. Today methods exist that can tackle both large numbers of electrons and the effects of relativity, but even these modern methods utilize approximations that decrease the accuracy of these calculations. CAST theoreticians are working directly with experimentalists to obtain the data needed to benchmark true, four-component, Dirac-Fock calculations on actinide molecules and materials. These efforts represent the state-of-the-art and are being led by researchers at LANL, SUNY-Buffalo, FSU, and the University of Manitoba.

Solution Chemistry. The solution chemistry of actinides is complex because these metal ions possess high coordination numbers, rapid ligand exchange kinetics, and there is a lack of even a rudimentary understanding of the origin of ligand preferences by actinide ions. In general, it is not currently understood whether enthalpic or entropic factors control the selectivity of separations processes. To solve these issues, efforts are underway in CAST that focus on quantifying *f*-element-ligand interactions both experimentally and computationally. These data are then used to fine-tune these interactions and achieve maximize selectivity. In addition to these more classical studies, CAST researchers are also advancing photochemical separations that capitalize on the narrow line-width of the *f*-element absorption features to selectivity transform ligands and enact separations simply using light from LEDS. Additionally, researchers at LANL, Purdue, UPenn, FSU, and FIU are utilizing X-ray spectroscopic methods to probe the electronic structure and speciation relevant to these separations processes. These spectroscopic measurements allow for a deeper understanding of actinide bonding and are used to benchmark the theoretical and simulation efforts.

Center for Actinide Science & Technology (CAST)	
Florida State University	Thomas Albrecht-Schmitt (Director), Susan Latturner, Kenneth Hanson, Eugene DePrince, David Hobart (Director of Operations)
Florida International University	Ines Triay (Deputy Director), Christopher Dares, Konstantinos Kavallieratos
National High Magnetic Field Laboratory	David Graf, Ryan Baumbach
Lawrence Berkeley National Laboratory	John Gibson
Purdue University	Susanne Bart
Los Alamos National Laboratory	Stosh Kozimor, Andrew Gaunt, Enrique Batista, Ping Yang
University of Pennsylvania	Eric Schelter
SUNY Buffalo	Jochen Autschbach
University of Manitoba	Georg Schreckenbach

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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
Class: 2014 – 2022

***Mission Statement**: To develop and harness a deep knowledge base in the characterization, prediction, and control of acid-gas interactions with a broad class of materials to accelerate materials discovery in acid gas separations, conversion, and utilization.*

Acid gases are ubiquitous in energy applications ranging from fuel and flue gas treatment and conversion of hydrocarbons to selective catalytic reduction of NOx and natural gas upgrading. The integrated research carried out by UNCAGE-ME (Phase I) allowed us to systematically identify materials features, common across classes of structures in some cases, that dictate how these materials behave in the presence of and/or selectively interact with acid gases. Having built this framework, the Center now seeks to harness this foundational knowledge through modeling, machine learning, and directed synthesis to accelerate materials design for acid-gas related energy technologies.

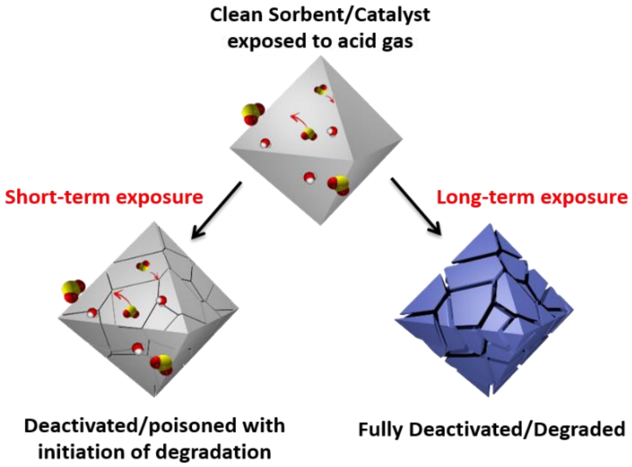


Figure 1: Changes induced in sorbents/catalysts upon short- and long-term exposure to acid gases. Materials are typically treated as passive in these environments, but can change dramatically upon exposure to SO_x, NO_x, and H_2S.

Materials-focused research for separations and catalysis often considers the materials as being passive in their process environment. In contrast, evolution of materials under working conditions due to exposure to acid gases is the norm rather than an exception. In Phase II of UNCAGE-ME, we will combine *in situ* molecular spectroscopic studies of both the surface functionalities and bulk structures of materials relevant to catalysis and separations of acid gases under conditions relevant to complex environments. Experiments will be combined with complementary data analytics and multi-scale computational and theoretical modeling of acid gas interactions with solids for targeted materials design.

Objectives

UNCAGE-ME seeks 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. With our discoveries on acid gas tolerance and reactivity of materials in hand, machine learning and data analytics can now be introduced as powerful tools for elevating these unique data sets to the level of real prediction. Coupled with in situ techniques and molecular modeling, UNCAGEME is now uniquely positioned to exploit systematic data on acid gas tolerance and reactivity to achieve an unprecedented level of design, prediction, and control of catalysts, membranes, and sorbents. Insights gained by the multi-investigator, multidisciplinary teams will allow us to achieve the following long-term, 4-Year Goals set forth for the Center:

1. Develop a deep knowledge base of structure-property relationships for acid gas interactions with sorbents, membranes, and catalysts in complex mixtures applicable to broad classes of materials.

2. Leverage machine learning techniques to enable fundamental predictions of materials interacting with complex gas environments on long time scales.
3. Harness computational techniques for directing the design and synthesis of catalysts, membranes, and sorbents with targeted selectivity and reactivity.
4. Accelerate materials discovery for acid-gas separations, conversion, and utilization through the development and use of an integrated set of design tools to predict material stability and lifetime in the presence of acid gases.

Center Research Team and Scientific Organization

In Phase II of UNCAGE-ME, the Center's research framework will include cross-cutting themes to drive materials discovery for predicting targeted chemistries, porosities, and defects, and the development of self-healing materials with a focus on regeneration methods. The original mission of UNCAGE-ME will continue to motivate the Center's research. New and ambitious research goals that leverage the success of our Phase I work while bringing new, powerful research capabilities to bear will drive the Center's work.

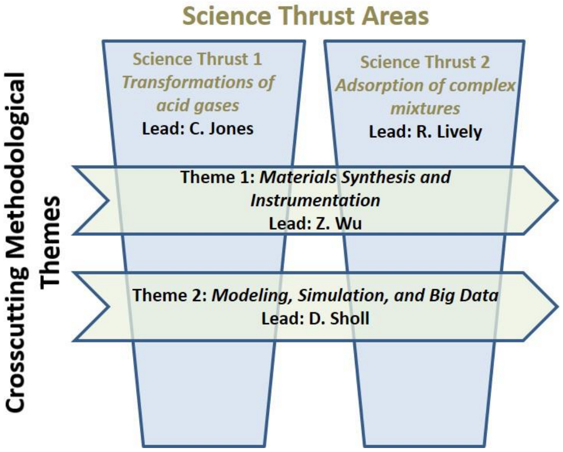


Figure 2: Overview of research thrusts/themes.

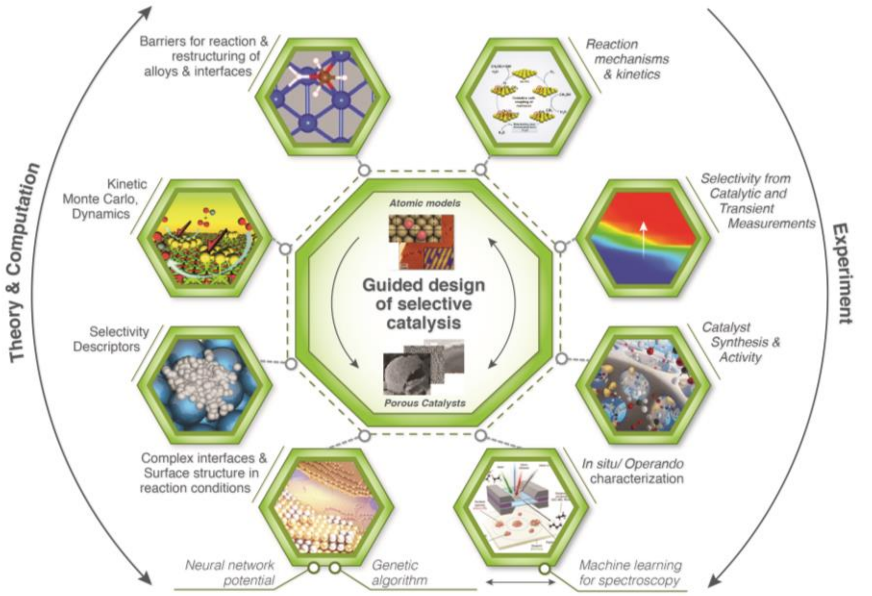
An overview of Science Thrusts and cross cutting Methodological Themes for Phase II of UNCAGE-ME is illustrated in Fig. 2. These Thrusts and Themes will work in an interconnected manner. The Science Thrusts focus on two broad strategies for developing the fundamental science for understanding and control of materials in acid gas mixtures relevant to two types of processes: (i) transformations of acid gases and (ii) adsorption of complex mixtures. The Methodological Themes are integrated across the Science Thrusts to assimilate the Center's results to predict high-performing materials, direct synthesis activities, and ultimately provide a robust, holistic design strategy that can be used more broadly by others in the field.

Center for Understanding and Control of Acid Gas-Induced Evolution of Materials for Energy (UNCAGE-ME)	
Georgia Institute of Technology	Krista Walton (Director), Christopher Jones, David Sholl, Ryan Lively, Sankar Nair, Rampi Ramprasad
Oak Ridge National Laboratory	Katharine Page, Bobby Sumpter, Zili Wu
Sandia National Laboratories	Jeffrey Greathouse, Tina Nenoff
University of Alabama	David Dixon
Lehigh University	Jonas Baltrusaitis, Israel Wachs
Pennsylvania State University	Susan Sinnott
Washington University St. Louis	Sophia Hayes
University of Wisconsin	JR Schmidt

Contact: Krista Walton, Director, walton@efrc.gatech.edu
404-894-5254, http://efrc.gatech.edu

Integrated Mesoscale Architectures for Sustainable Catalysis (IMASC)
EFRC Director: Cynthia M. Friend
Lead Institution: Harvard University
Class: 2014 – 2022

***Mission Statement**: To establish design principles for highly selective catalytic transformations driven by nanoporous dilute alloys.*



Scheme 1: Schematic of approach to design of selective catalysts in IMASC. A powerful combination of experiment and theory will be brought to bear on understanding key catalytic synthesis reactions on model systems and on functioning porous catalysts as a means of improving selectivity.

The Center for Integrated Mesoscale Architectures for Sustainable Catalysis (IMASC), an Energy Frontier Research Center, unites best-in-class researchers from universities and national laboratories to perform collaborative, innovative fundamental research at the intersection of surface chemistry and physics to transform how catalysts are designed. The integrated, Center-wide effort, built on a foundation of a well-managed matrix of projects and expertise, will enable us to reach ambitious strategic goals through innovative approaches and aggressive effort from a clear set of unified principles, fully committed collaborations and synergistic partnerships. The Center-wide goal is to develop the ability to improve catalytic selectivity by quantitatively scaling from model studies to catalytic conditions using advanced experiment and theory (Scheme 1). Improving reaction selectivity is extremely important because of the potential for increasing the efficiency of chemical production, which accounts for nearly 25% of energy use worldwide and which relies heavily on heterogeneous catalysis.

Heterogeneous catalytic processes are extremely complex, requiring optimization of factors across multiple scales of length, time, pressure and temperature. Hence, catalyst development requires the confluence of materials synthesis, mechanistic surface chemistry, and reaction kinetics. Complex metal/oxide interfaces are often present and may play an important synergistic role in the catalysis. Further, since the materials are often affected by the reaction environment, pre- and on-stream activation and optimization of performance is required. IMASC will capitalize on recent advances in theory and experiment to address these issues and thereby improve the efficiency of key catalytic processes.

The IMASC mission will be achieved by addressing several key scientific questions aimed at establishing principles for designing highly selective catalytic processes.

Key Scientific Questions:

1. How are the elementary steps in the reactive process affected by materials composition and structure?
2. What compositions, configurations, and structures of the alloy materials are optimal for high reactivity and high selectivity for specific reactions?
3. What is the effect of complex metal/metal oxide interfaces and how can they be controlled?
4. What is the state of the catalyst surface under reaction conditions and how does this affect selectivity and reactivity?

These questions will be addressed through the Center-wide projects that draw upon a range of experimental and theoretical expertise all managed in a matrix structure. Four cross-cutting methodological Expertise Areas will enable the research: EA1: Selectivity and Mechanism (Leads: Madix); EA2: Materials Synthesis (Lead: Biener); EA3: Theoretical Tools (Lead: Sautet); and, EA4: In Situ/Operando experiment (Lead: Stach).

Three **Center-wide projects** (thrusts) will focus IMASC efforts on critical catalytic transformations selected for their importance in chemical production.

A. Advancing the paradigm for mechanistic control of complex selective oxidation. (Lead: Madix)
B. Exploiting complex metal/oxide interfaces for challenging catalytic transformations. (Lead: Friend)
C. Directing selective hydrogenation: unsaturated oxygenates to nonthermodynamic products (Lead: Sautet)

The research in IMASC will culminate in a set of principles for designing dilute alloy catalysts capable of driving highly selective reactions. More broadly, an efficient methodology for understanding and improving catalytic processes will be established.

Integrated Mesoscale Architectures for Sustainable Catalysis (IMASC)	
Harvard University	Cynthia Friend (Director), Joanna Aizenberg, Boris Kozinsky, Robert Madix (Executive Committee Member)
UCLA	Philippe Sautet (Deputy Director)
Lawrence Livermore National Laboratory	Juergen Biener (Executive Committee Member)
University of Pennsylvania	Eric Stach (Executive Committee Member)
Tufts University	Maria Flytzani-Stephaopoulos, Charles Sykes
Brookhaven National Laboratory	Anibal Boscoboinik, Dario Stacchiola
Stony Brook University	Anatoly Frenkel
University of Florida	Jason Weaver
University College London	Michail Stamatakis

Contact: Leah DeFrancesco, Program Assistant, defrancesco@chemistry.harvard.edu
617-495-4198, https://efrc.harvard.edu/

Thermal Energy Transport under Irradiation (TETI)
EFRC Director: David Hurley
Lead Institution: Idaho National Laboratory
Class: 2018 – 2022

***Mission Statement**: To provide the foundational work necessary to accurately model and ultimately control electron- and phonon-mediated thermal transport in 5f-electron materials in extreme irradiation environments.*

To efficiently capture the energy of the nuclear bond, advanced nuclear reactor concepts aim to use fuels that must withstand unprecedented temperature and radiation extremes. In these advanced fuels, thermal energy transport under irradiation is directly related to reactor efficiency as well as reactor safety and is arguably one of the most important material properties. The science of thermal transport in nuclear fuel under irradiation is a grand challenge due to both computational and experimental complexities. In addition to accurately treating strong spin-orbit coupling of 5*f* electrons, the computation challenge also includes how to accurately model the formation of defects along with their influence on thermal transport.

The Center for Thermal Energy Transport under Irradiation ("Center" or "TETI") brings together an internationally recognized, multi-institutional team of experimentalist and computational materials theorists to develop a comprehensive, atom-to-mesoscale understanding of phonon and electron transport in advanced nuclear fuels. The Center takes a broad aim with two thrust areas: (1) phonon transport in advanced oxide fuels and (2) electron transport in advanced metallic fuels. The model fuels studied within the Center will be single crystal and polycrystalline thorium oxide and thorium oxide doped with uranium and the binary uranium-zirconium (UZr) system. This challenge is organized around a carefully constructed set of *science questions* that seek to close key knowledge gaps related to thermal energy transport in 5*f*-electron materials:

1. What is the impact of 5*f* electrons on phonon and electron structure in $Th_{1-x}U_xO_2$ and U-Zr alloys?
2. How do intrinsic and irradiation-induced defects self-organize in $Th_{1-x}U_xO_2$ and U-Zr alloys, and what are their impacts on electron and phonon scattering?
3. What are the collective effects of defects, defect ordering, and defect supersaturation on thermal transport $Th_{1-x}U_xO_2$ and U-Zr alloys?

Understanding electron- and phonon-transport characteristics from knowledge of chemistry / structure will have a far-reaching impact on materials development. Examples include utilization of defect organization under irradiation to improve thermal transport in advanced nuclear fuels, tailoring electron scattering in textured nanocrystals to enhance thermal conductivity of metal interconnects, simultaneously controlling electron and phonon transport in thermoelectric devices, and harnessing strong spin-orbit coupling to realize new paradigms for quantum materials.

Tackling the computational complexity is a far-reaching challenge. At the atomistic scale, the approach will involve using density functional theory plus dynamic mean field theory (DFT+DMFT) to understand the role of 5*f* electrons on phonon and electron transport, defect formation, and electron scattering mechanisms. At the mesoscopic-length scale, thermodynamic modeling and molecular dynamics will be used to understand defect interaction and evolution as well as phonon-scattering mechanisms. Also at this scale the Boltzmann transport equation (BTE) will be used to model transport.

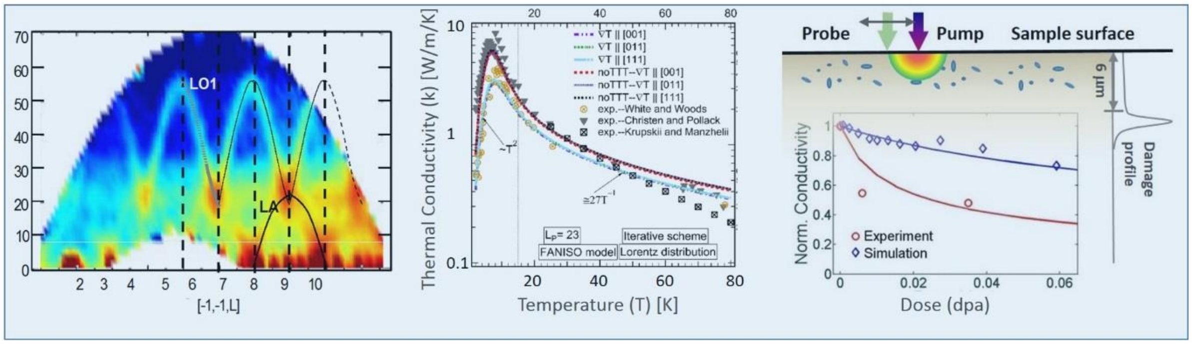


Figure. 1. *Left*: Dynamic structure of UO_2 measured along Q-[-1,-1,L] at 400k using the ARCS instrument at the Spallation Neutron Source, Oak Ridge National Laboratory. *Middle*: Simulation of the thermal conductivity of solid argon using the Boltzmann Transport Equation approach. *Right*: Time-resolved thermal wave measurement of conductivity in the plateau region of proton-irradiated UO_2 compared to MD simulation. These studies will be generalized to include both phonon and electron transport to investigate thermal transport in the $Th_{1-x}U_xO_2$ and UZr systems.

This modeling approach will be complemented by a well-defined set of electron- and phonon-structure measurements and transport measurements on ion-irradiated model fuels having well-characterized microstructures. Inelastic neutron scattering will be used to measure phonon dispersion and lifetime. Angularly resolved photoemission spectroscopy, and low-temperature magnetic field measurements (de Haas-van Alphen and Shubnikov-de Haas) will be used to obtain the electronic structure of metallic fuels. Thermal wave microscopy combined with coherent acoustic wave spectroscopy will be used to make spatially resolved thermal transport measurements across isolated grain boundaries and the damage plateau in ion-irradiated samples. The influence of supersaturation of point defects will be obtained by making first-of-its-kind in-reactor measurements of thermal conductivity.

The Center for Thermal Energy Transport under Irradiation (TETI)	
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Institute for Quantum Matter (IQM)

EFRC Director: Collin Broholm
Lead Institution: Johns Hopkins University
Class: 2018 – 2022

Mission Statement*: To realize, understand, and control revolutionary quantum materials and structures where quantum effects such as entanglement and coherence find collective macroscopic manifestations.*

The discovery and characterization of quantum materials is one of the grand challenges of twenty-first century physical science. Developed though a deep understanding of their underlying physics, this new class of materials could play an important role in extending the information technology revolution and confronting the unprecedented growth in global energy needs. But despite many important advances, quantum materials continue to present deep fundamental challenges. How can we predict and control the collective properties of 10^{23} electrons in a solid and what novel quantum dominated states of matter and electronic properties may ensue?

With a distinct focus on the discovery and understanding of new materials and artificial structures, the Institute for Quantum Matter Energy Frontier Research Center (IQM) will realize materials and structures where quantum effects such as entanglement and coherence find collective macroscopic manifestations. We shall expose, understand, and control the corresponding physical properties and explore their potential for energy relevant technologies. To accomplish this IQM comprises a collaborative team spanning the materials-by-design triad of materials discovery and synthesis (single crystals and thin films), advanced experimentation (neutron scattering, optical spectroscopies, transport, ultrasound, and high magnetic field techniques) and theory (analytical and numerical). IQM will focus on the discovery of four specific types of quantum matter that have not previously been realized:

Topological magnetic semimetals. While topologically protected 3D Weyl semimetals with linearly dispersing bulk excitations have been documented for inversion symmetry breaking systems, Weyl semimetals induced by collective magnetic order remain hypothetical to date. We shall design, synthesize, and characterize materials with the goal of realizing magnetic Weyl fermions borne of correlations. In related work we will also investigate a new class of materials with quadratic band touchings – the Luttinger semimetal state. Such systems are predicted to be generically strongly interacting and show non-Fermi liquid correlations.

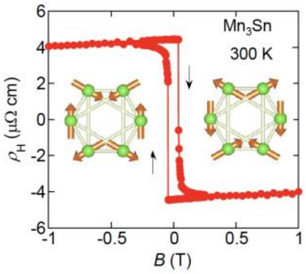

Fig. 1: Hall resistivity at room T in Mn_3Sn with spin configurations.

Quantum spin liquids. In a quantum spin liquid, quantum fluctuations destabilize magnetic order, even at T=0 K. There are now exactly solvable models with spin-liquid ground states and candidate materials to explore the underlying principles but definitive experimental evidence for a quantum spin liquid and demonstration of its topology remain elusive. IQM will develop new materials and spectroscopies to document emergent fractionalized excitations in quantum spin liquids – including photons and quantum Dirac monopoles – and utilize defects to characterize the underlying quantum spin liquid state.

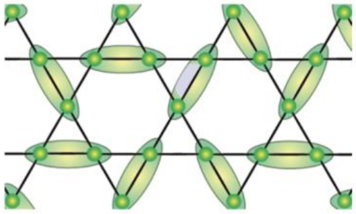

Fig. 2: A valence-bond configuration on a 2-dimensional kagome lattice. In a quantum spin liquid valence bonds resonate.

Monopole superconductors. The single particle energy gap in the density of states of a conventional superconductor is described by an analytical Cooper-pair wavefunction $\Delta(\mathbf{k})$ that can be classified in terms spherical harmonics. Superconductivity in an inversion symmetric Weyl semimetal is fundamentally different because the monopolar source of Berry curvature associated with a Weyl point within a Fermi-surface implies the phase of $\Delta(\mathbf{k})$ is no longer globally defined so the traditional classification scheme collapses. IQM will design, synthesize, and characterize materials and nano-scale structures to achieve and document a physical realization of monopole superconductivity.

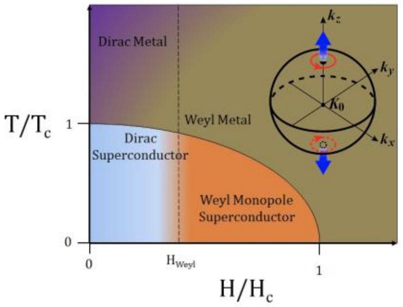

Fig. 3: The monopole superconductor has vorticity and may be realized in a Weyl semimetal.

Axion insulators. Topological insulators (TI) are unique states of matter that – despite an insulating bulk – harbor topologically protected surface states. We shall realize a related interacting magnetic state, the axion insulator. This is a theoretically proposed, but heretofore unrealized state of matter that is similar to a TI in possessing band inversion. However, axion insulators break time reversal symmetry and exhibit a large intrinsic magnetoelectric response that is quantized when inversion symmetry or another select point symmetry is preserved. Such systems are 3D analogs of the 2D quantum anomalous Hall systems and should show a quantized Kerr rotation for the inversion symmetric case and a measurable (and large) dc magnetoelectric response for the inversion symmetry broken state.

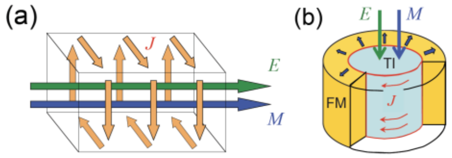

Fig. 4: Connection between magneto-electric coupling and surface current (a). TIs and axion insulators have a quantized surface anomalous hall effect. For TIs the surface states must be gapped by a ferromagnetic cladding layer (b) but this occurs spontaneously in axion insulators.

While each topic presents distinct challenges, there are also deep intellectual connections and cross-cutting methods and techniques so that successes in one area advances others. The unique physical properties of quantum materials present opportunities for breakthrough applications in energy and information. IQM is driven by the fundamental challenges that electronic correlations and topology present but as our understanding of quantum materials matures, we shall also bring their application potential into focus.

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Center for Novel Pathways to Quantum Coherence in Materials (NPQC)
EFRC Director: Joel Moore
Lead Institution: Lawrence Berkeley National Laboratory
Class: 2018 – 2022

Mission Statement*: To expand dramatically our understanding and control of coherence in solids by building on recent discoveries in quantum materials along with advances in experimental and computational techniques.*

The fundamental importance of the two-level system or qubit was recognized in the early days of quantum mechanics. An obvious way to maintain the quantum coherence of a qubit is to embed it in a perfect low-temperature vacuum. However, this is not the only way; a two-level system in a defect state in a solid (Figure 1), or a superposition of interband states at one value of momentum in an ideal crystal, can maintain quantum coherence for a remarkably long time even at room temperature. To fully exploit the potential of quantum-based sensing, communication, and computation, we must find new pathways to protect and use quantum coherence in solid-state environments that are closer to ambient temperatures.

This includes developing the power to manipulate coherence involving *many* two-level systems in realistic solid-state environments. New pathways will also come from applying advanced tools and concepts based on coherence to understand complex materials that could eventually provide alternate qubit, sensing, or optical technologies. Success in these basic goals requires an integrated approach via the EFRC and will provide new material approaches to a variety of quantum information science challenges.

Advances in quantum materials, including two-dimensional materials and topological materials, lead to remarkable new kinds of defects in both real space and momentum space. Major outcomes of this Center will include new approaches to solid-state quantum sensing and quantum spectroscopy, controllable crossovers between coherent and incoherent transport, and understanding the unconventional properties of a variety of new materials classes. These have the potential to open new frontiers in quantum information science, electronics, and optics. The work in this EFRC is aligned with BES Basic Research Needs for quantum materials, transformative experimental tools, and synthesis science.

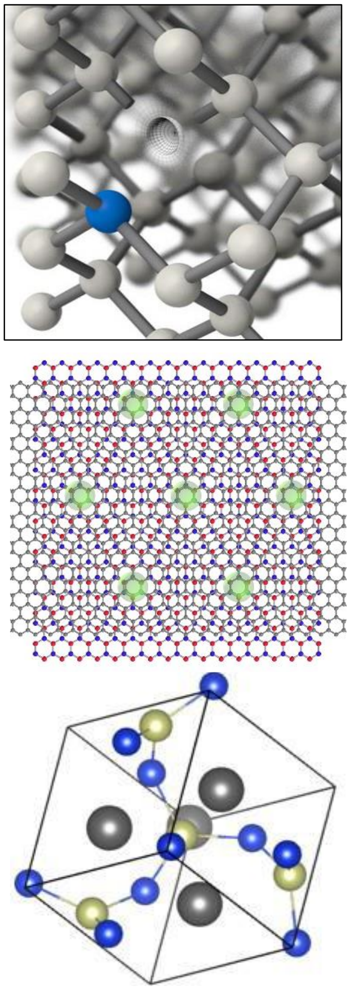

Figure 1. Illustrations of research directions for the three thrusts of this EFRC. *From top to bottom*: a nitrogen-vacancy (NV) center in the diamond lattice used for quantum sensing; 2D layers of trilayer graphene and boron nitride form a superconducting moiré superlattice; crystal structure of LaIrSi, an optically active topological material.

The Center conducts research in three major thrust areas:

Thrust 1 – Defects, disorder, and many-body entanglement for quantum spectroscopy: The ability to interrogate quantum materials and to measure their coherent properties is crucial for both the fundamental and applied sciences. Conventional wisdom holds that harnessing many-body entanglement can significantly enhance such quantum sensing technologies. The goal of this thrust is to theoretically predict, computationally optimize, and experimentally create, characterize, and develop novel defect-based quantum sensing platforms. In addition to the platforms themselves, we will investigate sensing protocols that utilize many-body interactions, non-equilibrium driving pulses, and quantum information inspired techniques (e.g., error correction) to improve sensor performance. In combination, these enhanced sensing methods will open new doors to directly image the nanoscale transport properties of heterostructures, the microscopic magnetic storage of information and the nonlinear optical response of quantum materials.

Thrust 2 – Quantum coherence in engineered surfaces: Two of the most significant achievements of the last decade in materials physics are deeper understanding of the importance of topological order in materials and dramatic improvement in our ability to engineer 2D materials with atomic precision. Thrust 2 is an outgrowth of these achievements, as it focuses on atomically-precise 2D material combinations that enable new types of topological and correlated quantum coherence. Research in this thrust explores atomically-engineered topological interfaces that promise to find new examples of topological protection and improve coherent transport to the point that it becomes technologically relevant. This thrust is also aimed at exploiting the new complex quantum states that are predicted to arise when 2D materials are combined in ways that take advantage of topological protection and many-body correlations.

Thrust 3 – Coherence and defects in correlated and topological materials: New materials with enhanced response functions have the potential to become transformative technologies: ultra-fast electronics on time-scales of quantum processes, ultra-sensitive sensors based on electronic phase transitions, and the ability to encode information at the nanoscale. The purpose of this thrust is to address a key challenge in realizing the potential of these materials: understanding the role of defects, disorder and heterogeneity in determining response functions. The goal is not only to mitigate their potential deleterious effects, but to investigate how they may be used to control and manipulate electronic properties.

Center for Novel Pathways to Quantum Coherence in Materials (NPQC)	
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Fundamental Understanding of Transport Under Reactor Extremes (FUTURE)
EFRC Director: Blas Pedro Uberuaga
Lead Institution: Los Alamos National Laboratory
Class: 2018 – 2022

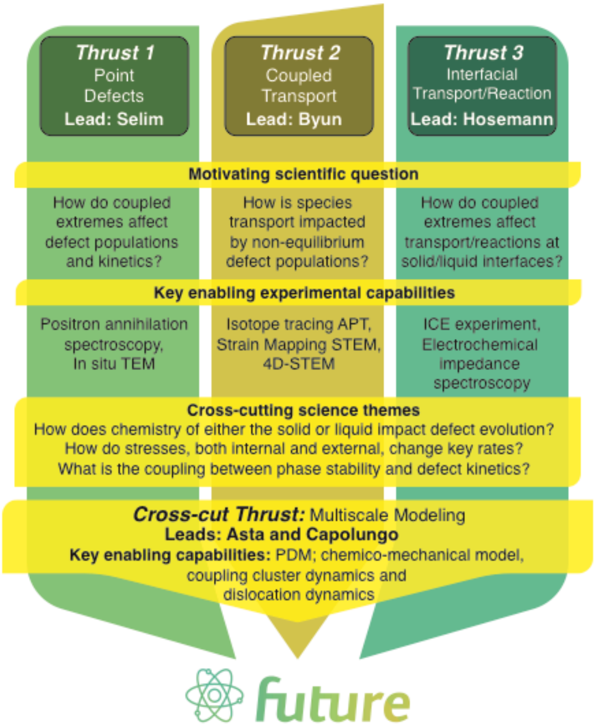
Mission Statement*: To understand how the coupled extremes of irradiation and corrosion work in synergy to modify the evolution of materials by coupling experiments and modeling that target fundamental mechanisms.*

Nuclear reactor environments are some of the most hostile and extreme built by humans. A multitude of harsh conditions exist simultaneously, all acting in concert to degrade the performance of the materials. These extremes include irradiation, temperature, stress, and corrosion. Irradiation damage itself is one of the greatest materials science challenges as it is truly multiscale, spanning from subatomic effects at the femtosecond time scale to macroscopic consequences for reactor components as large as the pressure vessel on the time scale of decades. Coupling irradiation with other harsh environments such as corrosion leads to an immense scientific challenge requiring a multidisciplinary team. We have assembled such a team in FUTURE.

In FUTURE, we target the response of materials to a combined corrosive and irradiation environment at relevant conditions of temperature and stress. Corrosion is driven by mass transport to and from reactive surfaces, across interfaces, and/or through protective scale layers. At the same time, the transport of species in the bulk material can lead to materials degradation. As the corrosion front advances, particularly when a new phase is formed via, for example, oxidation, stresses may build up that affect transport, altering both defect concentrations and mobility. On the other hand, radiation changes the concentrations and nature of the rate determining defects. That is, the defects that define corrosive behavior under thermal conditions may be irrelevant under irradiation. All of these defects will couple with elemental species intrinsic to the material and coming from the corrosive medium. It is critical to understand the coupling of irradiation-induced defects with elemental species in a corrosive environment to predict the response of the material in these coupled extremes.

By combining modeling and experiment, FUTURE targets these fundamental mechanisms. Our experimental campaign, broken into three Thrusts, focuses on three fundamental questions underlying the response of materials to coupled irradiation and corrosion extremes: what is the nature of the defects produced by irradiation, how do those defects couple with the various elemental species in the material to change the local chemical composition, and how is transport across interfaces modified by irradiation. We will advance unique experimental capabilities that target these questions. In the case of point defect generation during irradiation, we will complement in situ transmission electron microscopy (TEM) studies with in situ positron annihilation spectroscopy (PAS) to understand the nature of irradiation-induced defects. TEM is a work-horse in irradiation studies and is able to characterize and quantify the nature of large defect aggregates. PAS compliments this capability by being able to quantify point defects, such as vacancies and small vacancy clusters. We will design and install a PAS capability on LANL's existing ion beam lines to quantify these defects as irradiation is happening, as opposed to typical studies in which defect content is quantified after the fact.

These studies will be complemented by advanced microscopy characterization that examines how defects couple with elemental species in the material to modify overall evolution. In particular, we will use advanced environmental and 4D microscopy techniques to examine how microstructural features interact with defects and alloying elements to modify transport pathways. Uniquely, we will use isotopic atom probe tomography (APT) to determine how species such as oxygen migrate through the microstructure. By using isotopic labels, we will determine those regions in which transport is enhanced most significantly.



Finally, we will target transport across the liquid/solid interface. We will use electrochemical impedance spectroscopy (EIS) to quantify key reaction rates at these interfaces. We will also expand on a unique capability in which a coupled irradiation and corrosion environment is imposed directly on the material. This irradiation-corrosion experiment (ICE) has been used to study liquid metal corrosion. We will expand this to also target molten salt environments. This provides a direct and controlled experimental capability to understand how factors such as temperature and irradiation spectrum interact with the corrosive medium to modify the material properties.

All of this experimental activity is complemented by a cross-cutting modeling activity that aims to both elucidate the mechanisms observed in the experiments and develop a capability to predict the evolution of materials under these coupled extremes. The heart of this multi-scale effort is a new capability to be developed in FUTURE. This concurrent chemico-mechanical model, termed CD^3, combines cluster dynamics and dislocation dynamics to simulate the chemical evolution of a system in an evolving irradiated microstructure. The modeling framework will enable predictions that account for the microstructural evolution induced by irradiation that can be directly compared and validated against the experimental studies conducted in Thrusts 1-3.

Fundamental Understanding of Transport Under Reactor Extremes (FUTURE)	
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Pacific Northwest National Laboratory	Danny Edwards, Daniel Schreiber (Thrust Leader), Timothy Lach, Sandra Taylor
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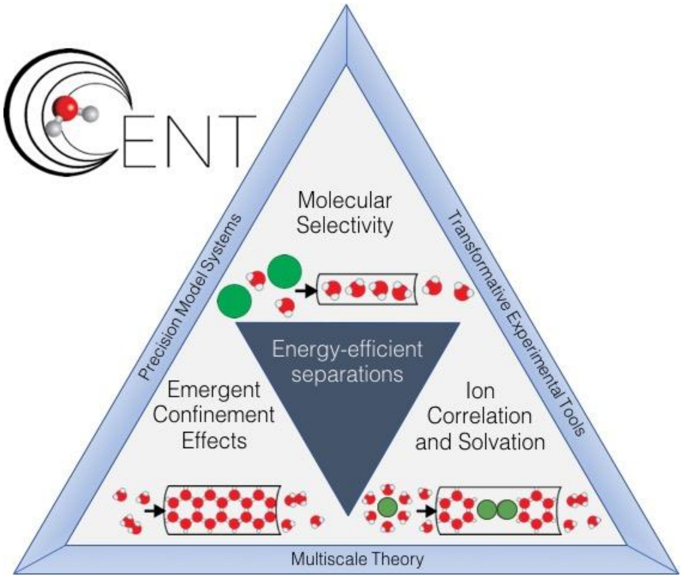
Center for Enhanced Nanofluidic Transport (CENT)
EFRC Director: Michael S. Strano
Lead Institution: Massachusetts Institute of Technology
Class: 2018 – 2022

***Mission Statement**: To address emerging and compelling gaps in our knowledge of fluid flow and molecular transport in single digit nanopores and establish the scientific foundation for developing transformative molecular separation technologies impacting the Water-Energy Nexus.*

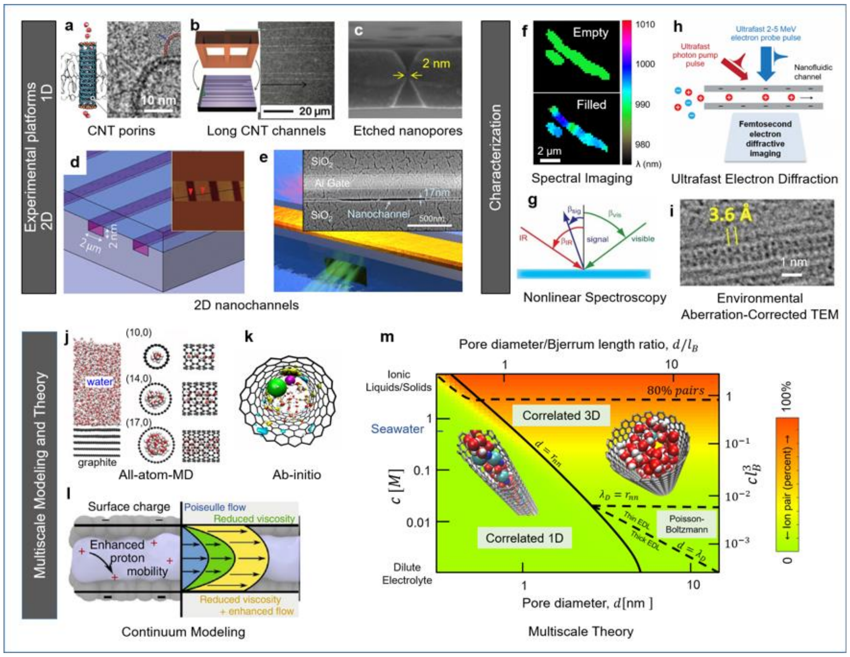


CENT will establish the scientific foundation for transformative molecular separation technologies based on SDNs to impact the Water-Energy Nexus.

Not all nanopores are created equal. By definition, all have characteristic diameters or conduit widths between approximately 1 and 100 nm. However, experiments indicate that the narrowest of such pores, Single Digit Nanopores (SDNs), defined as those with less than 10 nm diameters, display surprising behaviors resulting in extraordinary transport efficiency and selectivity These studies expose critical gaps in our understanding of nanoscale hydrodynamics, molecular sieving, fluidic structure and thermodynamics. Examples of these gaps include the observation of slip flow enhancement - that the narrowest of nanopores counter-intuitively demonstrate higher mass transport rates; evidence of non-Gibbs-Thomson phase behavior - that fluid phase boundaries in SDN are remarkably distorted from their bulk fluid counterparts; and highly non-linear, correlative effects in ion transport through SDN not observed in even slightly larger nanopore dimensions. These and other gaps are, in turn, an opportunity to discover and understand fundamentally new mechanisms of molecular transport at the nanometer scale that may inspire a host of new technologies at the Energy-Water Nexus, from novel membranes for separations and water purification to new gas-permeable materials and energy storage devices.

The Center for Enhanced Nanofluidic Transport (CENT) addresses 7 key nanofluidics knowledge gaps: (1) Slip Flow Enhancement in SDNs; (2) Fluid Phase Transitions in SDNs; (3) Phase-Separation under Confinement; (4) Defects and their Outsized Impact on SDN Transport; (5) Correlated Transport; (6) Nanoscale Solvation; and (7) Charge Exclusion to Significantly Enhance Selectivity. CENT research program is organized into three distinct and interconnected Thrust Areas: Understanding Emergent Confinement Effects (Thrust 1), Quantifying Ion Correlation, Dissipation, and Solvation Phenomena (Thrust 2), and Engineering Selectivity for Chemical Separations (Thrust 3). Our approach is highly integrated and is augmented by the development of precision model systems, transformative experimental tools, and predictive, multiscale modeling and simulations.

CENT is focusing on experimental platforms and analytical tools to understand the physical effects of extreme confinement in SDNs. We will address solvation effects in aqueous media and in acetonitrile,



CENT pursues an integrated, synergetic approach for probing emergent phenomena involving water and ions under confinement. CENT research combines experimental platforms that create 1D and 2D confinement with multi-scale simulation tools that model these effects using large scale, all-atom MD and *ab initio* simulations.

ethanol, and acetone solvents, which are important in energy-efficient separations, as well as propylene carbonate and representative ionic liquids used in energy storage devices. We will also explore fundamentally new mechanisms of ionic and molecular selectivity that stem from extreme confinement. CENT will pioneer the development of high fidelity fabrication methods for as small as sub-1 nm SDNs with conduit lengths of angstroms to 1 mm, with specifically tailored surface and pore mouth chemistry. These model SDN systems will provide the scientific basis for developing next generation membrane materials. CENT will also expand the ability to incorporate controlled, single defects into SDNs as a novel perturbation method and explore the impact of pore defects on nanofluidic transport. CENT team will pioneer the use of environmental aberration-corrected TEM, high resolution cryo-EM, ultrafast electron diffraction imaging, single ion traps, and single defect spectroscopy to transform our capability in probing water and ion structure in nanopores. CENT's work will produce a comprehensive, definitive theory, capable of predicting the thermodynamics and transport properties of fluids in SDNs. This knowledge in turn will motivate and enable new separation mechanisms for chemical purification and manipulation that would enhance the nation's competitiveness in the technologies at the Energy-Water Nexus.

Center for Enhanced Nanofluidic Transport (CENT)	
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Lawrence Livermore National Laboratory	Aleksandr Noy (Thrust 1 Lead), Tuan Anh Pham, Eric Schwegler
University of California Irvine	Zuzanna Siwy (Thrust 2 Lead)
Yale University	Mark Reed (Thrust 3 Lead), Menachem Elimelech
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Center for Hybrid Organic Inorganic Semiconductors for Energy (CHOISE)
EFRC Director: Matthew C. Beard
Lead Institution: National Renewable Energy Laboratory
Class: 2018 – 2022

Mission Statement: *To enable unprecedented synthetic control over the emergent phenomena of spin, charge, and light-matter interactions, in tailored organic-inorganic perovskite-inspired systems for energy science.*

Hybrid organic/inorganic semiconductors (HOIS) offer tremendous opportunities to control fundamental properties that underpin energy technologies. While currently there are enormous worldwide efforts exploring, exploiting and improving a narrow class of HOIS (lead-halide perovskites, such as methylammonium lead iodide ($MAPbI_3$)), primarily for photovoltaic (PV) applications, an opportunity exists to transcend this initial focus on PV research and seek deeper understanding and control of their fundamental properties. Inherent in these unique hybrid systems is the dichotomy between organic/molecular moieties (quantum chemistry) and inorganic/extended systems (solid-state physics). As a result, they exhibit properties that are not solely a juxtaposition of the inorganic and organic sub-units, but are instead truly emergent phenomena, with the concomitant ability to control and design new properties by judicious *choice* of inorganic and organic components.

While emergent physical phenomena in HOIS result from their unique hybrid nature, *rules for rationally designing HOIS systems and the precise underlying chemical/physical mechanisms underlying the emergent properties remain under investigation*. Semiconductor physics has revolutionized our world in the past century due to investments in fundamental studies of quantum mechanics and solid-state physics. Unfortunately, this knowledge is insufficient to completely describe HOIS. Complex coupling among inorganic and organic components drive unprecedented (and often collective) dynamic phenomena involving light, matter, and energetic species such as spins, charge carriers, and phonons. Thus, we have a *unique* opportunity to develop the foundational knowledge of this new paradigm in energy sciences.

The prototypical HOIS has a perovskite structure with ABX_3 (Fig. 1) stoichiometry where A is the organic cation, such as methyl ammonium ($CH_3NH_3^+$), B is the metal cation, such as Pb, and X is a halide anion. Many opportunities exist for HOIS in non-perovskite structures or with use of non-typical A, B, and X species, which can tune structure-property characteristics.

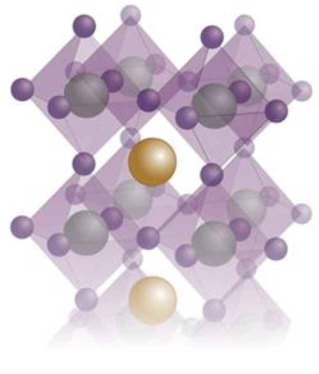

Figure 1(a) ABX_3. A (gold sphere) site is the organic component, B site is the center of the octahedra, X site are tips of the octahedra.

The ***Soft Nature*** is one distinguishing property that sets HOIS apart from conventional semiconductors, such as Si and GaAs. Softness corresponds to dynamical disorder within the lattice, whereby ions undergo large and reportedly strongly coupled motions (vibrations) about their average lattice positions. In 3D HOIS, the soft nature arises from weak noncovalent bonding, due to the large/anisotropic organic ion and the confluence of ionic bonding and anti-bonding coupling between lone pair *s* orbitals of the B cation and the halide *p* orbitals. The weak bonds are relatively easy to tilt, vibrate, and break, significantly influencing the structural, electronic, optical, and defect properties, and leading to emergent properties (Fig.2) that include: dynamic behavior, ferroelasticity, ferroelectricity,

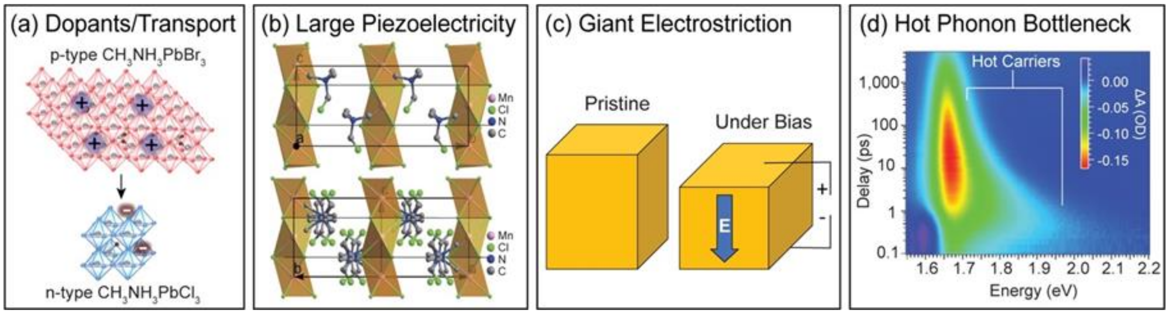

Figure 2. (a) Dopant compensation in $MAPbBr_3$. (b) Large piezoelectricity measured in $TMCM\text{-}MnCl_3$. (c) HOIS undergo giant electrostriction. (d) A hot phonon bottleneck dramatically slows carrier cooling.

electrostriction, photostriction, long lived hot-carriers, large polarons, broadband light emission, and low recombination rates. The presence of strong spin-orbit coupling (SOC) and breaking of inversion symmetry lead to other unique effects in HOIS, based on the effect known as ***Rashba Splitting***. Rashba-Splitting should have important impacts on key phenomena underpinning energy technologies, including: long spin-coherence times, low recombination rates, high photoluminescence yields and spin-to-charge conversion, among others.

Goals for CHOISE (Fig. 2) are:

- Uncover *design rules dictated by the distinct requirements of organic/inorganic chemistry,* which allow for high-precision control over composition, structural dimensionality, defect density, orientation, and conformation of organic molecules within HOIS.
- Control SOC and Rashba-splitting, so as to uncover and exploit the unique roles of the inorganic and organic sub-components in the presence of large SOC enabling *unprecedented control over the energetics, coherent transport, and dynamics of spin populations*.
- Elucidate and exploit structure/function relationships, both within the bulk and at HOIS surfaces and interfaces, that enable energy-efficient interconversions between light, excitons, spins, and charge carriers.
- Control the degree of 'softness' to discover how *charges couple to **static** and **dynamic** lattice properties,* enabling emergent phenomena such as ferroelasticity, ferro- and piezoelectricity, giant electrostriction, slow hot-carrier cooling, and controlled doping.

Center for Hybrid Organic Inorganic Semiconductors for Energy (CHOISE)	
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National Renewable Energy Laboratory	Matthew C. Beard, Joseph Luther, Joseph Berry, Kai Zhu, Jeffrey Blackburn, Jao van de Lagemaat
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University of Utah	Valy Vardeny, Sarah Li
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Center for Next Generation of Materials Design (CNGMD)
EFRC Director: William Tumas
Lead Institution: National Renewable Energy Laboratory
Class: 2014 – 2020

***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.*

To realize the *Next Generation of Materials Design*, we will couple first-principles theory with state-of-the-art high throughput and directed synthesis and characterization, including novel *in-situ* methods to understand and predict structure, properties, and phenomena at the molecular, nano, and meso scales. Our current work is focused on three key science questions: 1) How to quantify the synthetically accessible limit of metastability for a number of general synthetic approaches; 2) How to predict synthetic pathways to produce high-energy polymorphs; and 3) How functionality and synthetic pathways can be defined for new multinary oxides and nitrides originating from crystalline and amorphous precursors.

Accordingly, the goals of the CNGMD EFRC are:

***Goal 1: Design and discover new energy-relevant materials** with targeted functionalities by integrating theory, high-throughput computation, synthesis, and characterization.* This integrated approach to accelerating materials design and synthesis incorporates both multi-property design and inclusion of metastable materials.

***Goal 2: Incorporate metastable materials** into Materials by Design and establish an understanding of metastability including composition, and structure and formation energies for polymorphs and semiconductor alloys.* We will specifically investigate classes of metastability relevant to inorganic semiconductors for optoelectronic applications including polymorphism, semiconductor alloys, and thermochemically metastable compounds.

***Goal 3: Advance predictive synthesis science** for stable and metastable functional materials by coupling calculated energetics, intermediate structures, and reaction parameters with experimental observables and in situ measurements for 1) aqueous synthesis, 2) solid-state synthesis, and 3) synthesis from amorphous precursors.* 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 how the materials in Goal 2 can be achieved.

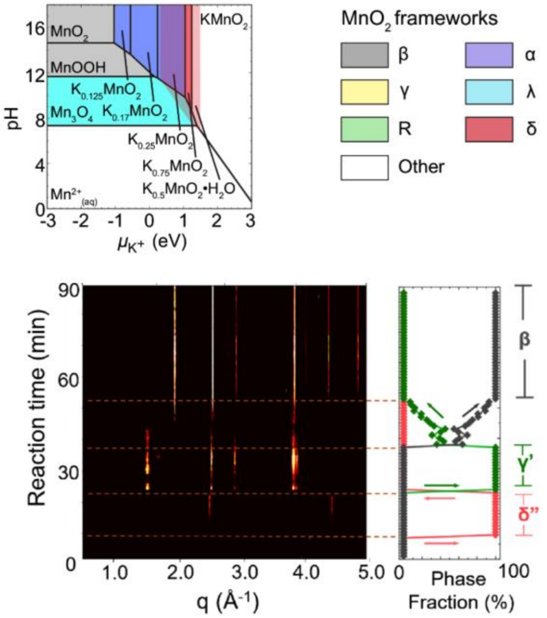

Fig. 1: Top - Predicted pathways for MnO$_2$ polymorph synthesis. Bottom – Phase formation during synthesis measured using *in situ* x-ray diffraction.

Goal 4: Promote and disseminate *the Next Generation of Materials Design to the broader materials science community*. This includes making our methodology and data accessible as well as organizing conference symposia and workshops.

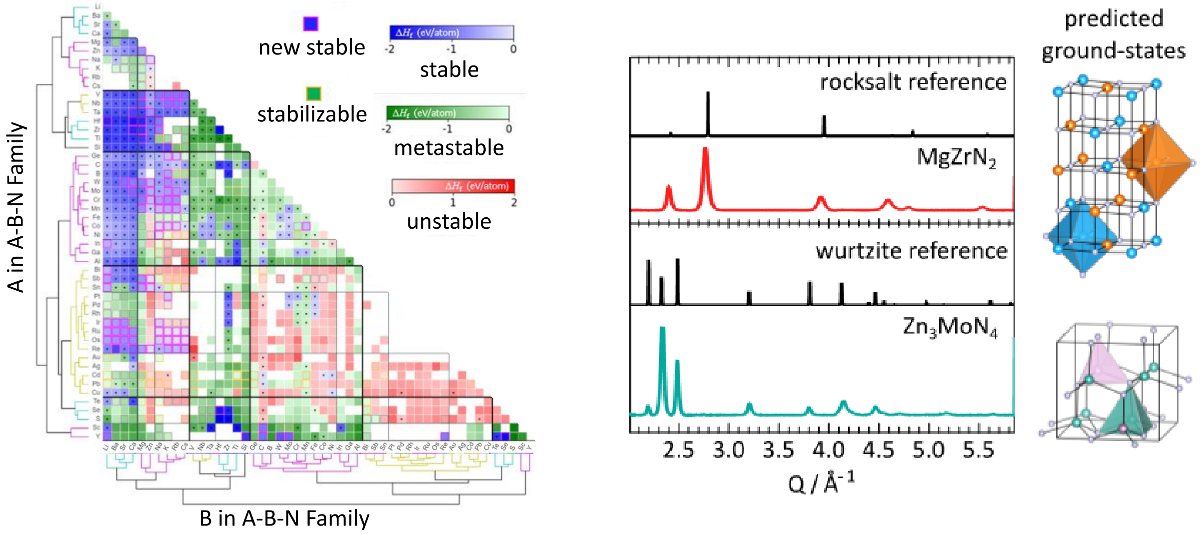

Fig. 2: Left - Map of the inorganic ternary metal nitrides colored by the thermodynamic stability. Right - Predictive synthesis of new ternary nitride functional materials.

Our materials focus is on semiconductors for solar energy conversion, solid-state lighting, power electronics and related technologies—all areas in need of transformative materials. Our current work focuses on two key thrust areas:

Thrust 1: Predictive Synthesis of Complex Metal Oxides will use aqueous and solution-phase synthesis as well as synthesis from amorphous precursor films. We will focus on controlling the synthesis of high energy polymorphs and functional ternary metal oxides.

Thrust 2: Nitride Materials will focus on understanding and controlling nitride metastable materials and metastable heterostructural nitride alloys.

Center for Next Generation of Materials Design (CNGMD)	
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Lawrence Berkeley National Laboratory	Gerbrand Ceder (Chief Theorist), Kristin Persson
SLAC National Accelerator Laboratory	Michael Toney
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Center for Bio-Inspired Energy Science (CBES)
EFRC Director: Samuel I. Stupp
Lead Institution: Northwestern University
Class: 2009 – 2022

***Mission Statement**: To develop the next frontier in soft materials relevant to energy challenges by designing structures that emulate functions we see in biological systems.*

Soft materials are normally composed fully or partially of organic matter and the best examples are polymers, which have had an enormous impact in energy relevant technologies, particularly energy savings in transportation, manufacturing, infrastructure, and construction, among others. The next challenge is to learn how to synthesize soft materials with the capacity to interconvert energy forms, for example the way muscles convert chemical to mechanical energy in living organisms, or the way plant leaves optimize light to chemical energy conversion in a resourceful way to synthesize chemicals. Our vision is that basic science research in this area can lead to artificial materials that rival living ones in the remarkable and useful ways they manage energy. Our proposed research program specifically tackles the next big challenges in synthetic design of soft materials, namely learning how to encode in them molecularly the ability to transduce energy forms and even move autonomously in ways that are characteristic of "living matter". We approach this enormous bio-inspired challenge through bottom-up chemical design and synthesis combined with top-down engineering strategies, computation, and theory to create novel functional systems. The goal is to develop through basic science new opportunities around the concept of "robotic soft matter", denoting its autonomous ability to rapidly perform mechanical, optical, or chemical tasks with only small inputs of electrical energy and without the use of complex hardware. Equally important is learning to create "photosynthetic matter", which requires systems structured holistically to enable efficient chemical production using visible light. Our targets to create robotic and photosynthetic soft matter are extremely relevant to future modalities in manufacturing and chemical production, two of the greatest users of energy. Our cross-disciplinary investigations focus on bio-inspired research in the following areas.

Multi-Scale Synthesis of Artificial Muscles (inspired by energy transduction in muscles)

Muscles are composed of soft materials that have fast mechanical responses to chemical inputs. We have already demonstrated the ability to create soft robotic materials that mechanically respond to thermal or light inputs, but at relatively slow response speeds. Our aim now is to explore the upper limits of response speed in these systems, to make them fast-acting. Our approach will include both molecular synthesis and top-down architectures of the structures.

Magnetic Morphogenesis (inspired by biological development)

During biological development, protein signals mediate the morphogenesis of large collections of cells into specific shapes that create functionality. We aim to create the same level of control over synthetic soft matter using magnetic fields that actuate motion at nano, micron, and millimeter scales. We plan to use morphogenesis to induce directed locomotion and actuation.

Autonomous Soft Microrobots **(inspired by living cells)**

Living cells navigate complex environments to perform diverse functions by integrating the capabilities of sensing, actuating, computing, and communicating. Similarly, we envision developing shape-shifting "microrobots" that move autonomously and adapt their motions in response to both environmental cues and interparticle signals. Microrobots with encoded functions are potentially desirable for distributed sensing or healing/repair tasks in energy-relevant materials such as battery electrolytes, polymer membranes, and catalysts.

Hierarchical Structure-Mediate Photocatalysis **(inspired by the spatial organization of functional molecules in biological systems)**

Biological photosynthesis occurs in highly structured environments where the position and order of the components play a key role in the overall efficiency of multiple energy and electron transfer steps. Focusing on fundamental questions regarding the design of photocatalytic materials with bio-inspired spatial organization, we will explore how hierarchically assembled synthetic materials can be used to emulate the light-driven reactions found in biological systems.

Mechanical Enhancement of Photocatalysis **(inspired by leaves)**

Leaves use mechanical forces to physically rearrange their chloroplasts to control photosynthetic output. We recently discovered surprising changes in the visible light absorption spectra and photocatalytic activity of hydrogels which depend on subtle changes in supramolecular packing. We will explore the potential impact of mechanical forces on the performance and control of soft matter encoded for photocatalytic activity and the possibility of accessing non-equilibrium photocatalytic states using mechanical energy.

Center for Bio-Inspired Energy Science (CBES)	
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Center for Light Energy Activated Redox Processes (LEAP)
EFRC Director: Michael R. Wasielewski
Lead Institution: Northwestern University
Class: 2009 – 2020

***Mission Statement**: To develop the fundamental scientific understanding needed to use efficient light-driven multi-electron redox processes to power energy-demanding chemistry.*

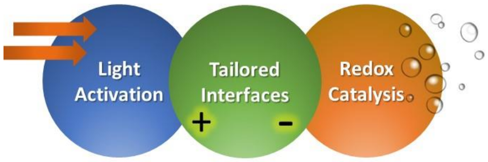

Fig 1. Synergistic LEAP Research Thrusts.

Understanding energy-demanding redox reactions is important because they are the basis of processes used on a global scale to provide materials, fuels, chemicals and food. Making these processes highly efficient will have a major positive impact on both the US and global economy. Leveraging the substantial advances made by the ANSER Center EFRC, LEAP Center research addressing our mission during the next two years is organized into three interwoven Thrusts with closely integrated approaches and team synergies:

- **Thrust 1. Powering Redox Processes.** Will produce charges at potentials required to drive energy-demanding redox reactions with temporal and spatial precision using unconventional organic/inorganic semiconductors.
- **Thrust 2. Tailored Interfaces and Hierarchical Assemblies.** Will tailor interfaces between **Thrust 1** semiconductors and **Thrust 3** redox catalysts using hierarchical assemblies to control charge flow to power the catalysts.
- **Thrust 3. Redox Catalysts for Energy-Demanding Reactions.** Will design, synthesize, and characterize molecules and materials with catalytic metal centers that will utilize charges provided by **Thrust 1** semiconductors to perform energy-intensive reactions, such as H_2O splitting, CO_2 and N_2 reduction, and C-C bond formation.

Thrust 1: Powering Redox Processes.

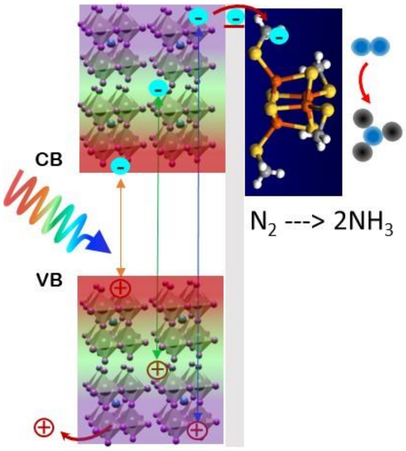

Fig 2. Schematic of interaction of a PS with an N_2 reduction catalyst, and hot-carrier extraction to catalytic sites via control of structure and recombination dynamics.

*The goal of LEAP **Thrust 1** is to leverage emerging classes of unconventional semiconductors to create charges at potentials sufficient to drive the catalysts described in **Thrust 3** to carry out energy-demanding redox processes. The interfacial strategies of LEAP **Thrust 2** will be used to couple the semiconductors to the catalysts and stabilize the overall system.* Achieving this goal will involve four types of closely related materials: organic semiconductors (Oss), perovskite semiconductors (PSs), quantum dots (QDs), and 2D dichalcogenides. These new semiconductors have recently astounded the energy conversion community with their long excited-state lifetimes, excellent charge mobilities, large photocurrents, broad optical absorption, chemical versatility, and the advancing stability. Specifically, OSs, especially as bulk-heterojunction structures, hybrid organic-inorganic halide PSs, QDs, and hybrid organic–2D inorganic materials have enabled major advances in fundamental understanding, many made by ANSER, which have resulted in greater extraction of photon energy with open-circuit voltages well above 1 V and fill factors near 80%. The approaches described here aim to achieve potential differences between extracted charges of up to 1.5-2.0 V to drive catalytic chemical reactions.

Thrust 2: Tailored Interfaces and Hierarchical Assemblies.

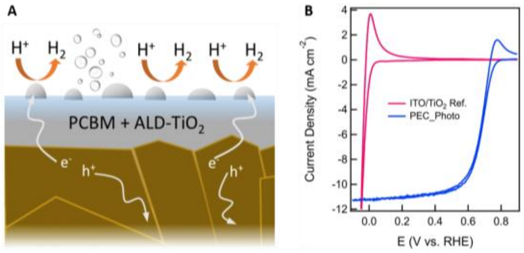

Fig. 3. A) Schematic of perovskite photocathode for proton reduction. **B)** I-V characteristics of ITO/PEDOT:PSS/halide-perovskite/PCBM/TiO_2/Pt in strong acid under simulated solar illumination vs an ITO/TiO_2/Pt control.

*The goal of LEAP **Thrust 2** is to understanding the interplay between materials and molecular chemistry to temporally and spatially control the flow of photo-generated charges to productive redox catalytic centers, while stabilizing all system components under conditions that are far from thermodynamic and chemical equilibrium.* To achieve catalysis of light-driven thermodynamically uphill reactions, **Thrust 2** aims to create and understand interfaces and hierarchical structures (Figure 3) that contain structurally and chemically well-defined linkers between light absorbers, redox catalysts, and electrodes, and that are robust under operating conditions. These structures will efficiently channel charge carriers into desired reaction pathways, minimize unproductive pathways and side-products, avoid high energy intermediates, and create states that store energy for triggered release.

Thrust 3: Redox Catalysts for Energy-Demanding Reactions.

*The goal of LEAP Thrust 3 is to design catalysts for efficient electron-driven chemical transformations—catalysts that can be interfaced/integrated (**Thrust 2**) with unconventional semiconductors (**Thrust 1**). Achieving this goal will involve understanding how light-powered molecular and heterogeneous catalysts, and their designed environments beyond the binding site, accomplish electron-driven, chemoselective transformations involving important molecular substrates such as CO_2 and N_2. These studies will yield efficient new catalysts, instructive new reaction mechanisms and pathways, and fundamental insights into the challenge of manipulating reaction networks in complex environments.*

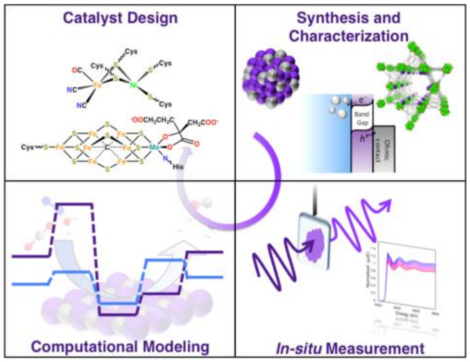

Fig. 4. Computational and experimental design, synthesis, characterization, and functional mechanistic assessment of atomically precise catalysts (**Thrust 3**) for integration (**Thrust 2**) with designed photoelectrodes (**Thrust 1**).

Thrust 3's approach (Figure 4) will integrate theory and experiment to: a) design catalysts; b) develop new ways to synthesize desired catalysts; c) characterize designed catalysts *in operando*; d) evaluate complex mechanisms; and e) measure catalyst chemoselectivity and energy efficiency against theory. The precept is that mechanistic understanding of specific reactions will yield transferable design rules for existing and potentially new classes of catalysts and reactions.

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Fluid Interface Reactions, Structures and Transport (FIRST)

EFRC Director: Sheng Dai
Lead Institution: Oak Ridge National Laboratory
Class: 2009 – 2022

Mission Statement*: To achieve fundamental understanding and validated, predictive models of the atomistic origins of electrolyte and coupled electron transport under nanoconfinement that will enable transformative advances in capacitive electrical energy storage and other energy-relevant interfacial systems.*

The FIRST Center will develop accurate atomic to nanoscale computational models to replace our inadequate phenomenological understanding of *ion transport* that limits power density in capacitive systems, where the need for ultrahigh surface area dictates nanoporous/nanotextured materials. Under nanoconfinement, the reduced dimensionality, interfacial interactions, surface charge, and disruption of ion-solvent and correlated ion-ion interactions are expected to profoundly affect transport**.** Reduction/oxidation (Redox) reactions introduce additional energy storage and it is critical to understand the coupling of ion transport with local electron transfers in order to control *fast and reversible surface redox processes* under confinement. Determining the system-scale environments needed to promote fast electrolyte ion transport and rapid surface redox reactions will enable new paradigms for *simultaneous high electrical power and energy*. To achieve our mission, summarized in **Figure 1.** we will build on our progress in understanding fluid-solid interface (FSI) structures and reactivity in electrical energy storage, by addressing the following critical scientific knowledge gaps:

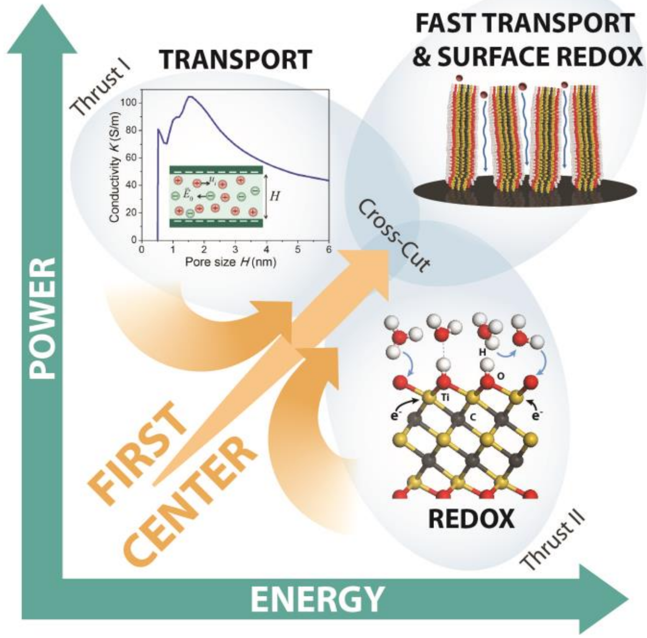

Figure 1. Conceptual summary of the FIRST Center mission and goals. Inset figures, counterclockwise from to top left: Classical Density Functional Theory predicts above-bulk conductivity of an ionic-liquid-like electrolyte confined in a charged graphene-like nanopore under an external electrical field[1]; Fast surface redox reaction at metal centers in Ti_3C_2 MXene is linked with fast proton transfer and surface adsorption within interlayers in aqueous electrolytes[2]; Alignment of MXene flakes perpendicular to current collector facilitates ion and electron transport to achieve simultaneous high pseudocapacitive power and energy density[3].

(1) *What ultimately controls the transport properties of nanoconfined electrolytes?*
(2) *How does correlated ion transport depend on pore confinement and electrolyte concentration?*
(3) *Is the transport of electrolyte ions coupled with electron transport and surface charge evolution in nanostructured electrodes?*
(4) *How are rates and mechanisms of pseudocapacitive redox processes controlled by nanoconfinement and interfacial ion transport?*

Fig. 1 relates our organizational structure to the mission and goals of the FIRST Center. Our research is conducted in two synergistic research **Thrusts**: (**I**) *Electrolyte Transport Properties and Confinement Effects* and (**II**) *Coupled Electrolyte Ion and Electron Transport in Redox-Active Media.* In **Thrust I**, we determine what precisely controls nano- to meso-scale diffusive and convective transport of electrolyte species within nanopores and related nanoconfined interlayer spaces to understand what ultimately limits how fast an electrochemical supercapacitor can charge and discharge. In **Thrust II**, we draw upon insights gained in **Thrust I** to determine the controls of electrolyte species transport and nanoconfinement on fast and reversible surface redox reactions needed to understand pseudocapacitive storage. A **Cross-Cutting Theme** uses the fundamental insights gained in **Thrusts I** and **II** to understand how novel pseudocapacitive system architectures facilitate *Simultaneous High Power and High Energy*. By integrating novel experimental and computational approaches, we will learn how to predictively utilize local and mesoscale environments in nanoporous/nanotextured media over multiple length scales to cooperatively enhance ion and/or electron transport, which are central to achieving high energy and power delivery. Our research directly addresses Priority Research Directions identified in the DOE Office of Basic Energy Sciences reports on *Next Generation Electrical Energy Storage*[4] and *Energy and Water*[5]. Success in achieving our mission and goals will also lay the foundations for advances in chemical separations, desalination, heterogeneous catalysis, waste isolation and many other energy relevant fluid-solid interface systems.

References Cited:

[1]C. Lian, A. Gallegos, H.L. Liu, J.Z. Wu, Non-scaling behavior of electroosmotic flow in voltage-gated nanopores, *Phys. Chem. Chem. Phys, 19*:450-457 (**2017**).

[2]C. Zhan, M. Naguib, M. Lukatskaya, P.R.C. Kent, Y. Gogotsi, D.-E. Jiang, Understanding the MXene pseudocapacitance, *J. Phys. Chem. Lett., 9*:1223-1228 (2018).

[3]Y. Xia, T.S. Mathis, M.-Q. Zhao, B. Anasori, A. Dang, Z.H. Zhou, H. Cho, Y. Gogotsi, S. Yang, *Nature, 557*:409-412 (**2018**).

[4]G. Crabtree, G. Rublof, E. Takeuchi, *Report of the Basic Research Needs Workshop on Next Generation Electrical Energy Storage*, Mar. 27-29, 2017.

[5]M. Tirell, S. Hubbard, D. Sholl, *Report of the Basic Research Needs Workshop on Energy and Water*, Jan. 4-6, 2017.

Fluid Interface Reactions, Structures and Transport (FIRST)	
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Energy Dissipation to Defect Evolution (EDDE)
EFRC Director: Yanwen Zhang
Lead Institution: Oak Ridge National Laboratory
Class: 2014 – 2020

Mission Statement*: To understand how extreme chemical complexity can be exploited to control energy dissipation and defect evolution under equilibrium and non-equilibrium conditions and to guide the development of radiation-tolerant alloys with unique magnetic and thermal properties.*

Means of improving the performance of structural materials have been intensively investigated for many decades. Solid solution strengthening, traditionally achieved by alloying minor elements into pure metals or conventional alloys, is one of the most widely used methods to attain specific desirable properties, including radiation tolerance. It has long been recognized that alloys with specific compositions exhibit enhanced radiation resistance; however, it remains unclear how the atomic-level structure and chemistry affect defect formation and damage evolution during irradiation. This knowledge gap has been a roadblock to the development and implementation of future-generation energy technologies.

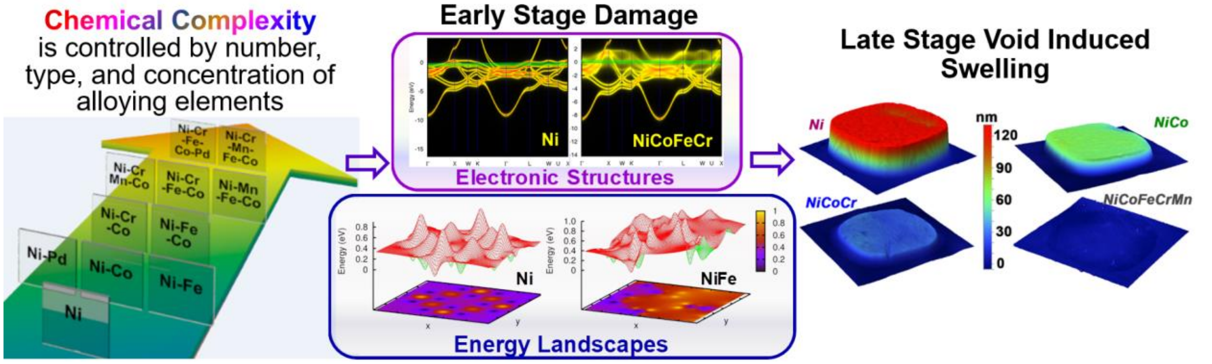

Fig. 1. EDDE aims to understand and tailor chemical complexity in model alloys (subset of SP-CSAs shown on left), which represents a powerful tool for dramatically modifying electronic and atomic properties (middle), and ultimately enhancing radiation tolerance (right) by reducing the rate of Energy Dissipation (Thrust 1) and controlling Defect Evolution (Thrust 2).

The predictive discovery and guided design of advanced materials with targeted functionalities will be key to enabling modern technologies, especially for future nuclear energy applications. A new class of alloys—Single–Phase Concentrated Solid–solution Alloys (SP-CSAs) composed of multiple elements, all at high concentrations—exhibit unexpected and exceptional properties, e.g., robust phase stability, ultrahigh low-temperature toughness, high-temperature strength, superparamagnetism, superconductivity, and importantly, suppressed irradiation-induced damage accumulation and void formation. In the EDDE Center, we hypothesize that by understanding and optimizing the chemical complexity both locally and globally in SP-CSAs (Fig. 1 left; Zhang, et al., *Current Opinion in Solid State and Materials Science* **2017**, 21, 221–237), energy dissipation and defect evolution can be tailored upon individual radiation events to encourage defect annealing within the picosecond time scale (Fig. 1 middle; Zhao, et al., *Physical Review Materials* **2018**, 2, 013602; Zhang, et al., *Nature Communications* **2015**, 6, 8736), and ultimately suppress damage accumulation under prolonged high-dose irradiation (Fig. 1 right; Lu, et al., *Nature Communications* **2016**, 7, 13564). We will understand and exploit the ability to tailor the chemical complexity of SP-SCAs, to thereby control energy dissipation via energy carriers that transport charge, heat, and spin (Fig. 1 middle top) and to control defect evolution resulting from kinetic energy transfer and mass transport during and after radiation events (Fig. 1 middle bottom), e.g., collision cascades and defect migration. The EDDE Center will study the extreme chemical complexity that arises from intrinsic elemental disorder in SP-CSAs to fundamentally understand an alloy's performance in extreme environments and reveal design principles for the predictive discovery and guided synthesis of new alloys with targeted functionalities, well beyond radiation tolerance.

EDDE's research is structured around two intertwined research Thrusts implicit in the Center's name: Energy Dissipation (Thrust 1) aims to understand the complex interactions among the energy carriers (electrons, phonons, and magnons) that dissipate elastic and inelastic energy deposited in a material; Defect Evolution (Thrust 2) aims to control the energy landscapes and tailor atomic transport processes to impact both defect production during collisional energy transfer in the femtosecond to picosecond time frame and damage evolution over longer time scales. The EDDE Center has extensive experience in growing high-quality, large, single crystals with targeted compositions; collaborative execution of well-defined irradiation experiments and microstructural characterization (including *in situ* ion beam analysis techniques, ultrafast science, and high-resolution microscopy/spectroscopy); world-leading theoretical and modeling expertise; and development of high-performance computer codes that extend the size and complexity of systems amenable for theory, modeling, and high-throughput discovery. The study of SP-CSAs through a combination of deep theoretical understanding, state-of-the-art computational approaches, and cutting-edge synthesis and experimental methods will enable us to elucidate the mechanisms controlling defect dynamics and irradiation performance of SP-CSAs based on fundamental energy scattering mechanisms. The synergy between the EDDE Center's team members within ORNL and with its partner institutions will allow EDDE research to capitalize on the team's outstanding strengths, thereby achieving rapid scientific advances.

The development and study of unique SP-CSAs in the EDDE Center have prompted new questions that challenge established theories and models currently applicable to conventional (dilute) alloys. Our overarching goal is to provide comprehensive knowledge of how the chemical complexity of SP-CSAs manifests and ultimately controls energy dissipation and defect evolution under extreme conditions. The success of the EDDE Center will provide a foundation that will enable breakthroughs in the design of revolutionary new materials and will present unforeseen opportunities for materials discovery.

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Center for Performance and Design of Nuclear Waste Forms and Containers (WastePD)
EFRC Director: Gerald Frankel
Lead Institution: The Ohio State University
Class: 2016 – 2020

***Mission Statement**: to understand the fundamental mechanisms of waste form performance and apply that understanding to design new waste forms with improved performance.*

The report of a 2015 DOE workshop on Basic Research Needs for Environmental Management described the need for "new materials for long-term storage of nuclear waste, including waste forms." The main performance criterion for waste forms, and the primary consideration for the design of new ones, is their ability to isolate and immobilize the radionuclides by withstanding corrosion in a repository environment over geologic time scales. WastePD is focused on this topic, specifically the environmental degradation mechanisms and the science underlying the design and performance prediction of new corrosion-resistant waste forms and canister materials.

Nuclear waste will eventually be placed in a mined repository in different forms, including three classes of solid matter: glasses, crystalline ceramics, and metals, and all three can degrade during exposure to the environment. WastePD consists of three thrusts focused on glass, ceramic, and metal corrosion, as shown in the figure. The thrusts are tied together by synergistic activities, intersectional topics of study, and an aligning framework.

Glasses and crystalline ceramics are often considered to be nonreactive and corrosion resistant, but they do degrade. When exposed to water, glass reacts to form a thin alteration layer on its surface that protects the glass and slows the rate of attack. In some environments, however, this protective layer becomes unstable and the glass exhibits an accelerated rate of attack. The details of the exact nature and stability of the protective layer are not known, so the transition to a non-protective condition cannot be predicted. As a result, current repository performance models take a conservative approach by assuming that glass will corrode at an accelerated rate rather than the slow passive rate. A better understanding of the glass corrosion process could lead to improved glass waste forms, support more robust repository models, and influence the repository design.

Crystalline ceramic waste forms can also release the incorporated radionuclides during interaction with the environment through matrix dissolution, selective leaching, radionuclide diffusion and surface reaction. An alteration phase may also form on the surface of ceramic waste forms to protect against further corrosion. Furthermore, many ceramic hosts under consideration for nuclear waste are semi-conductive and therefore can undergo transformative electrochemical reactions in aggressive environments. The environmental degradation of ceramic waste forms has not been studied in detail and little is known about the phenomena involved.

By their nature, metals are often susceptible to environmental degradation by electrochemical processes, although extremely corrosion resistant alloys (CRAs) like stainless steels (SS) have been developed by the judicious combination of alloying elements and prescribed processing. Like glass and ceramics, CRAs are protected by the spontaneous formation of a very thin surface oxide layer. However, such nm-thick films can break down and lead to accelerated forms of corrosion. The design of CRAs has in the past been based on empirical knowledge, which is not useful when considering new classes of alloys such as High Entropy

Alloys (HEAs). New approaches shown to be successful for computational design of materials have not been extended to corrosion resistance. Thus, it is clear that there are unifying concepts in the corrosion of the three materials classes of waste forms.

The design and long-term performance prediction of all materials, including glass, ceramics and metals, is currently empirical, based on correlations, experience, and intuition. The main goal of WastePD is to develop the Science of Environmental Degradation of Materials (SEDMat), with applicability to all three materials classes. SEDMat will enable a transition from empiricism to design based on calculable parameters, and ultimately to multiscale, multiphysics models that describe and predict the sub-processes of the overall corrosion phenomenon.

SEDMat will require enhanced fundamental understanding. For nuclear waste glass corrosion, it is critical to attain physical insights into the structure of the glass/solution interface and the behavior of reactive and inactive species present at that interface. Focus is on the formation of an amorphous, hydrated, porous, alumino-silicate layer on the glass and ceramic surfaces, and the roles of composition, structure and near-field environment. The corrosion of crystalline ceramics, in addition to short-range structural effects similar to glass, is also influenced by grain size and grain boundary structures. Different model ceramic systems, including apatite, hollandite, and perovskite are investigated as promising forms for incorporating I, Sr, and Cl fission products. The focus is on the interfacial behaviors across solid-solid and solid-liquid boundaries that can be closely linked with the ceramic waste form corrosion and stability under near field conditions. Metal HEAs, like multicomponent waste glasses, have almost limitless possible composition variations that enable fundamental material design concepts to be tested and validated.

WastePD is the first center ever created to address the corrosion of glass, crystalline ceramics and metals in a comprehensive and coordinated manner. The interactions between experts in the different materials create new approaches and understanding that would not have been possible otherwise. An example is how WastePD provides a unique synergistic platform for studying the interaction of corrosion behaviors between material classes. Glass or ceramic waste forms in a repository will be exposed to environments resulting from the corrosion of SS canisters. Studies of glass and ceramic wasteforms corroding in contact with SS are providing new insight into how SS corrosion products affect glass and ceramic corrosion and how the glass and ceramic corrosion products in turn influence SS corrosion.

Center for Performance and Design of Nuclear Waste Forms and Containers (WastePD)	
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Center for Molecular Electrocatalysis (CME)
EFRC Director: R. Morris Bullock
Lead Institution: Pacific Northwest National Laboratory
Class: 2009 – 2022

***Mission Statement**: To establish the fundamental principles needed for efficient interconversion of electrical energy and chemical bonds through precise control of electron and proton transfers.*

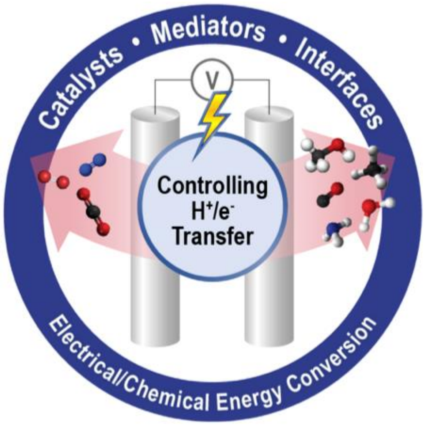

Figure 1. Efficient electrocatalysts require precise control of movement of protons and electrons.

Energy production in the United States is evolving rapidly, with growing contributions from wind and solar. The increasing generation of intermittent and off-peak electricity requires reliable energy storage (Fig. 1). Chemical bonds are ideal for energy storage because of the high energy density of chemical fuels, as well as the large scale at which fuels can be stored. Revolutionary solutions to this challenge require transformative fundamental discoveries in electrocatalysis and interfacial reactivity to achieve energy-efficient interconversion of electrical and chemical energy. We will pioneer a system-level approach to improve catalyst performance by simultaneously targeting three integrated goals: (1) *transcending scaling relationships*, (2) *enhancing catalysis using mediators*, and (3) *achieving molecular control of interfaces* (Fig. 2). The broad scope of these efforts will be addressed through closely coupled experimental and computational studies, grounded in thermochemical analysis and novel theoretical approaches.

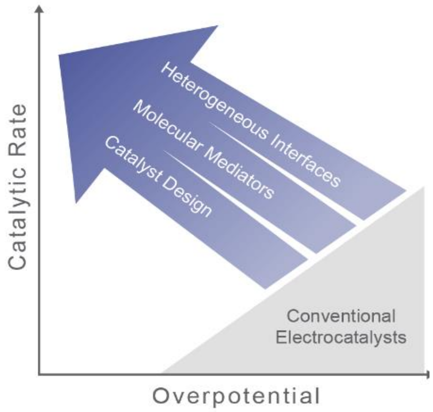

Figure 2. CME seeks to improve the rates, energy efficiencies, and selectivities through the design of catalyst systems that surpass the limitations suggested by conventional correlations between these parameters.

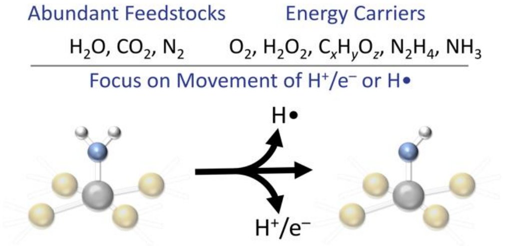

Figure 3. By focusing on the thermodynamics and kinetics of the movement of protons/electrons, CME will develop the principles needed to design catalysts with unprecedented performance.

The **Catalyst Design** research thrust seeks to transcend the limitations of conventional linear free-energy correlations between rate, overpotential, and selectivity by controlling the thermodynamics of proton-coupled electron transfer (PCET) reactions. (Fig. 3). We will show how the mechanistic characterization of catalytic reactions reveals not one, but multiple scaling relationships. Our work will contribute to the strategic design of new catalyst systems with dramatically improved performance. The **Molecular Mediators** research thrust will establish that electron-proton transfer mediators (EPTMs) provide an effective way to achieve "redox communication" between an electrode and a molecular or heterogeneous catalyst in bulk solution (Fig. 4). This use of EPTMs as "soluble electrodes" greatly expands the scope of electrode-driven

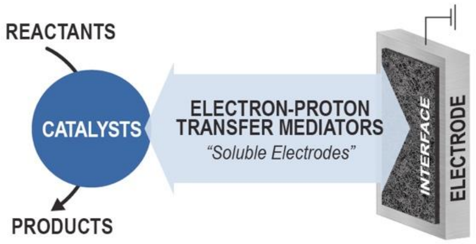

Figure 4. Mediated catalyst design is an emerging concept that will improve system performance by delivering H^+/e^- away from the electrode.

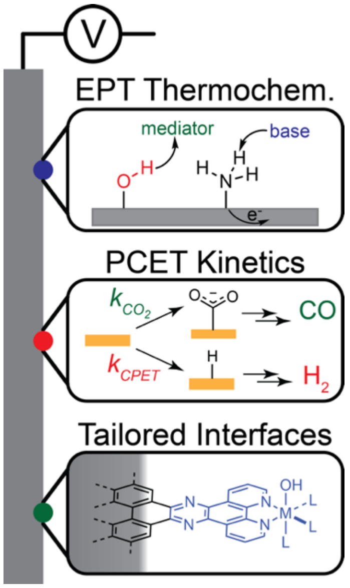

Figure 5. CME uses a molecular approach to tailor interface reactivity.

redox processes and provides the basis for dramatic improvements in the rates and/or overpotentials of these reactions. We intend to enhance electrocatalyst performance through cooperativity with tailored EPTMs and mediated electrocatalyst systems. The **Heterogeneous Interfaces** research thrust will address the challenge of controlling e^-/H^+ transfer steps at heterogeneous interfaces, achieving control through insights from molecular reactivity (Fig. 5). PCET at the surface of electrodes and heterogeneous catalysts is a crucial basic science frontier underlying diverse electrochemical energy transduction reactions. Our cross-cutting **theoretical and computational activities** are aimed at understanding quantitative design principles for efficient control of proton and electron movement. These computational efforts will guide the exploration and optimization of the complex free energy landscape of catalytic processes. Methodological developments will have potential impact beyond the research topics directly addressed in our work, advancing computational capabilities for electrochemical PCET and atomistic modeling of electrochemical processes at interfaces.

Fundamental principles developed through the proposed research will impact a range of scientific disciplines, as the bond-forming and bond-breaking reactions studied are applicable to many key questions in energy science. We envision a future energy landscape that has been radically transformed from the familiar long-standing practices, in which fuels will be produced at ambient temperature using electrocatalysts, starting from abundant molecules such as nitrogen, carbon dioxide, and water. The chemical energy in N-H or C-H bonds will be converted directly to electricity using fuel cells based on earth-abundant metals, obtaining high energy efficiency.

Center for Molecular Electrocatalysis (CME)	
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Yale University	Sharon Hammes-Schiffer (Deputy Director), James Mayer (Lead, Heterogeneous Interfaces Thrust)
University of Wisconsin – Madison	Shannon Stahl (Lead, Molecular Mediators Subtask), Thatcher Root
Massachusetts Institute of Technology	Yogesh Surendranath
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Interfacial Dynamics in Radioactive Environments and Materials (IDREAM)
EFRC Director: Sue Clark
Lead Institution: Pacific Northwest National Laboratory
Class: 2016 – 2020

Mission Statement*: To master molecular to mesoscale chemical and physical phenomena at interfaces in complex environments characterized by extremes in alkalinity and low-water activity, and driven far from equilibrium by ionizing (γ,β) radiation.*

IDREAM is an Energy Frontier Research Center (EFRC) conducting fundamental science to support innovations in processing high-level radioactive wastes (HLW). IDREAM facilitates the transformation of HLW processing by elucidating the basic chemistry and physics required to control and manipulate interfacial phenomena in extreme HLW environments (e.g., heterogeneous and chemically complex). This foundational knowledge is required to achieve ***IDREAM's mission to master molecular-to-mesoscale chemical and physical phenomena at interfaces in complex environments characterized by extremes in alkalinity and low-water activity, and driven far from equilibrium by ionizing (γ, β) radiation.*** Further, understanding these processes will enable prediction of waste aging over the many decades required to complete this difficult cleanup task. IDREAM activities focus on aluminum hydroxides and (oxy)hydroxides, as they are principal components of bulk waste materials to which a variety of other metal ions and radionuclides partition.

Through novel and highly integrated experimental, computational, and theoretical approaches, we propose to develop the ability to predict and control the critical physicochemical phenomena currently preventing the application of more efficient and cost-effective tank waste removal and processing strategies. Our work will span from the molecular scale (e.g., speciation and dynamics of key solution species), to interfacial dynamics (e.g., dissolution and precipitation rates and mechanisms), to particle scale (e.g., particle interactions), and will involve realistic conditions of extreme alkalinity, low water activity, and ionizing radiation. IDREAM will provide a scientific foundation for emergent phenomena in high-level radioactive waste (HLW) processing, such as dissolution rates, aging phenomena, and slurry behavior including adhesion, friction, gelation, and rheology.

As shown in the figure below, IDREAM is structured around three research thrusts and three cross-cutting thrusts:

Research Thrust 1 (RT1): **Molecular and Solution Processes**. Understand the roles of solvent dynamics, chemical reactivity, solute organization, and pre-nucleation species on overall molecular speciation in highly alkaline systems of concentrated electrolytes.

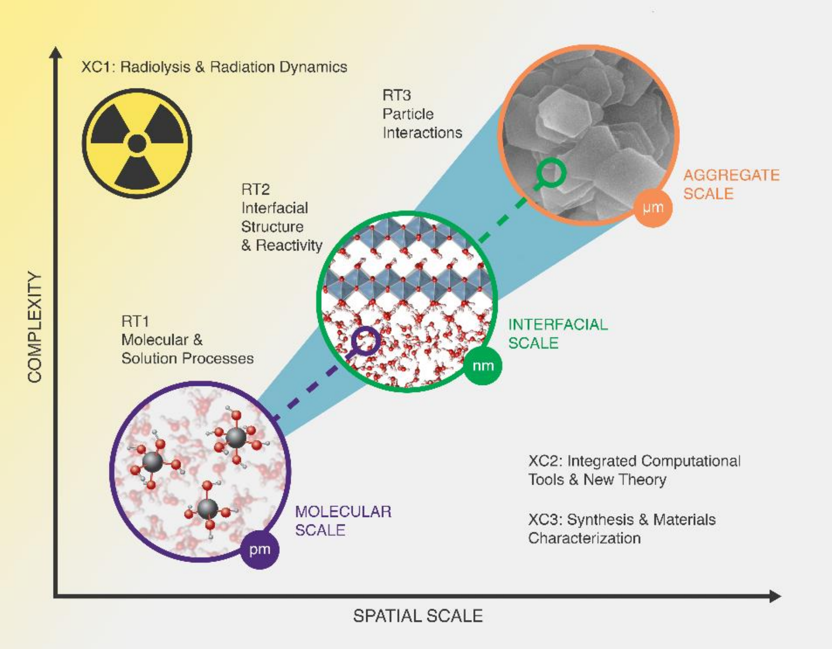
Research Thrust 2 (RT2): **Interfacial Structure and Reactivity.** Link fluid phase dynamics to nucleation, particle growth, and interfacial reactivity in highly alkaline systems of concentrated electrolytes.

Research Thrust 3 (RT3): **Particle Interactions**. Quantify the chemical and physical phenomena leading to hierarchical microstructures of aggregates, and other response dynamics that couple across scales.

Cross-Cutting Thrust 1 (XC1): **Radiolysis and Radiation Dynamics**. Quantify the role of β and γ radiation in driving molecular speciation and interfacial reactivity far from equilibrium.

Cross-Cutting Thrust 2 (XC2): **Integrated Computational Tools and New Theory Development.** Develop and apply integrated computational approaches within and across research thrusts to enable foundational, new theories in interfacial chemistry to emerge.

Cross-cutting Thrust 3 (XC3): Synthesis and Materials Characterization. Understand and control the synthesis of aluminum oxides and oxyhydroxides to design and produce materials with well-defined characteristics.



IDREAM has a central theme of aluminum chemistry because it's commanding role in the processing of high level radioactive wastes. It is a primary component of the bulk solids to which other metal ions and radionuclides are partitioned, and must be dissolved and separated from radioactive contaminants. This focus provides an integrating theme across the four Research Goals. The integrated disciplinary perspective of IDREAM will accelerate the transformative understanding of complex interfacial phenomena that are driven far from equilibrium. We will exploit a broad range of characterization tools and computational resources within our collaborating institutions and at DOE user facilities.

By better understanding chemical interactions across scales of time and space, we can explain poorly understood macroscale phenomena, such as waste stream rheology, slurry agglomeration, and control of precipitation. This will provide the foundation for accelerating the dissolution of the waste materials, its processing, and improving process systems. The knowledge gained in IDREAM will be broadly applicable to other complex heterogeneous processing problems related to materials and energy production.

Interfacial Dynamics in Radioactive Environments and Materials (IDREAM)	
Pacific Northwest National Laboratory	Sue Clark (Director), Carolyn Pearce, Kevin Rosso, Jaehun Chun, Greg Kimmel, Greg Schenter, Jim De Yoreo
Washington State University	Aurora Clark, (Deputy Director), Kerry Hipps
University of Washington	Xiaosong Li
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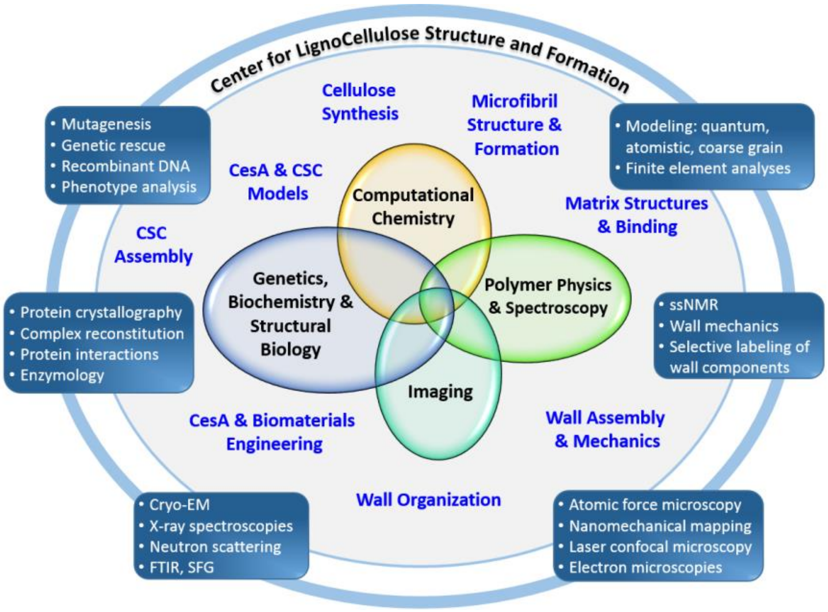
Center for Lignocellulose Structure and Formation (CLSF)
EFRC Director: Daniel J. Cosgrove
Lead Institution: Penn State University
Class: 2009 – 2022

Mission Statement: *To develop a nano- to meso-scale understanding of cellulosic cell walls, the energy-rich structural material in plants, and the physical mechanisms of wall assembly, forming the foundation for new technologies in sustainable energy and novel biomaterials.*

Plant cell walls - also known as cellulosic biomass or lignocellulose - are among the most complex, diverse and useful materials on Earth. These hierarchical structures represent an abundant and renewable source of valuable biomaterials and bioenergy, presenting untapped *transformative opportunities* for engineering them with new properties while simultaneously providing lessons on how to *mimic the nano-scale structure and means of assembly of these complex living materials* for synthesis of man-made materials with specific, tunable properties.



CLSF's research is at the nexus of physics, chemistry and biology and draws on expertise from diverse fields, diagrammed at right. Insights from our research will form the foundation for future efforts to optimize the structures and utility of plant cell walls, which are essential to plant life and comprise a large-scale source of renewable biomaterials and bioenergy.

CLSF goals in the current phase will build upon advances made in the previous funding period to:

1. Combine multiple state-of-the-art methods of electron microscopy with neutron and X-ray scattering, computational modeling and biochemistry to solve the structure and catalytic mechanism of plant cellulose synthases (CesAs) and native cellulose synthesis complexes (CSCs).

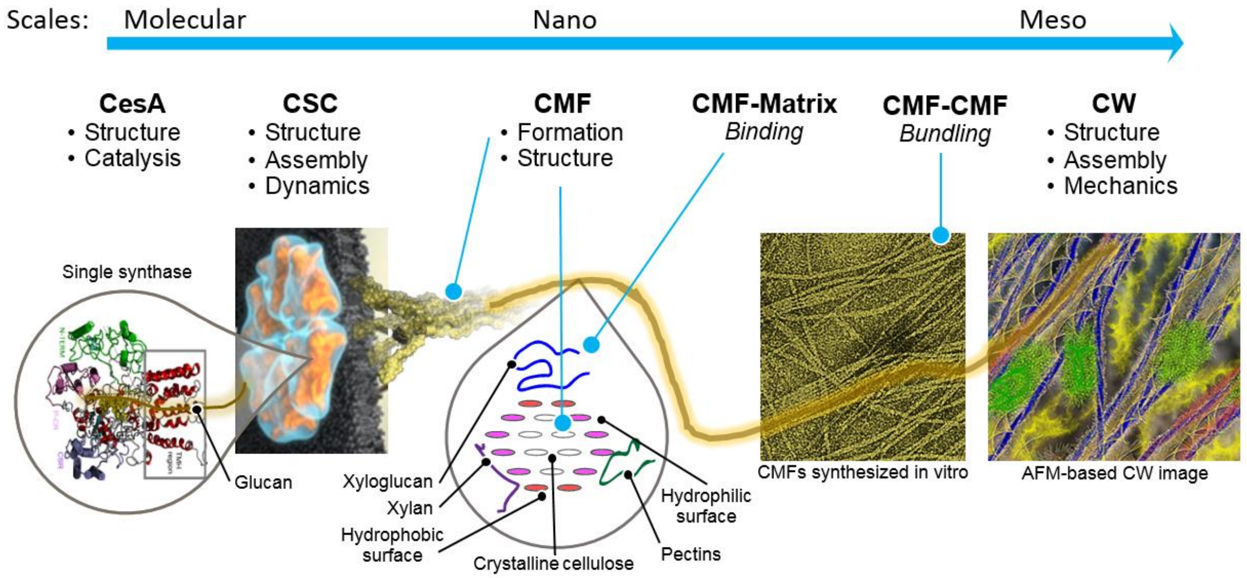
2. Manipulate active CesA assemblies *in vitro* and *in vivo* to learn how artificial and native CSCs are assembled and how cellulose microfibril structure depends on CSC structure. We will use these new experimental platforms to test computational models of CSC and cellulose microfibril assembly.

3. Develop new experimental and quantitative methods for assessing cellulose microfibril organization in cell walls and use them to uncover the physical mechanism(s) of microfibril bundling.

4. Extend newly-developed methods and results by CLSF to analyze the physical basis of microfibril-matrix interactions in cell walls with different matrix polymers and study the structural, physical and mechanical consequences of altering these interactions in primary and secondary cell walls.

5. Develop new biological systems (such as the growing Arabidopsis inflorescence stem and xylem-transdifferentiation in transgenic seedlings) to study the processes of microfibril bundling, primary cell wall assembly and maturation, and secondary cell wall formation.

These topics are linked to one another as illustrated graphically below:



Above: Research questions addressed by CLSF include the structure and kinetics of cellulose synthase (CesA); the structure and activity of the cellulose synthesis complex (CSC); cellulose microfibril (CMF) structure and CMF interactions with water, matrix polysaccharides and lignin.

These goals involve new teaming arrangements and development of novel approaches, experimental platforms and advanced instrumentation. The five goals will synergistically produce new insights for potential means to achieve control of man-made materials and for ways to tune cell walls for specific properties in the materials and energy fields. Overall success with even a subset of these goals will enable a quantum leap in understanding how plants assemble these complex hierarchical structures.

Center for Lignocellulose Structure and Formation (CLSF)	
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Oak Ridge National Laboratory	Hugh O’Neill
North Carolina State University	Candace Haigler, Yaroslova Yingling
Massachusetts Institute of Technology	Mei Hong
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Bioinspired Light-Escalated Chemistry (BioLEC)
EFRC Director: Gregory Scholes
Lead Institution: Princeton University
Class: 2018 – 2022

Mission Statement: *To employ light harvesting and advances in solar photochemistry to enable unprecedented photoinduced cross-coupling reactions that valorize abundant molecules.*

The energy input required to transform stable and abundant molecules to valuable products is greatly reduced by the use of catalysts. A fundamental aim in catalysis is to devise new ways to convert plentiful and unreactive molecules for energy-relevant applications. The research proposed for the BioLEC Energy Frontier Research Center (EFRC) will expand our understanding of fundamentals of solar photochemistry and photosynthetic systems to enable sophisticated cross-coupling chemistry powered by light, Figure 1. The resulting breakthroughs will yield energy-relevant chemicals, fuels, and materials. At the frontier of this endeavor, we aim to catalyze reactions that have prohibitive energy barriers for equilibrium chemistry—reactants are more stable than products. The reactions that we target are presently inconceivable using the leading edge of modern synthetic chemistry. Our approach is inspired by the way that nature combines the energy of multiple photons to ramp up redox capability beyond that achievable with the energy from a single photon. To succeed, BioLEC unites scientific communities that rarely interact—organic synthesis, structural and molecular biology, and physical chemistry.

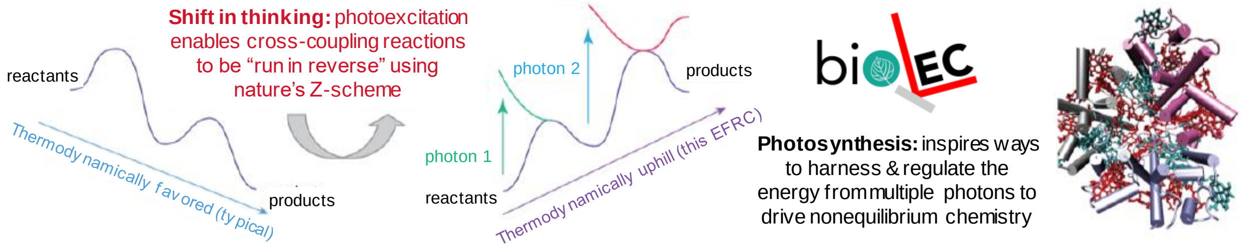

Figure 1. BioLEC's interdisciplinary approach will enable nonequilibrium chemistry.

The BioLEC EFRC strives to achieve a profound understanding of solar photochemistry and photosynthetic systems to enable photoinduced cross-coupling chemistry to create valuable chemicals, fuels, and materials. Our research approach to developing photocatalysis capable of creating valuable chemicals from abundant molecules is organized within four research thrusts. In Thrust A (Lead: Abigail Doyle), we develop new single-electron-transfer photoinduced cross-coupling systems, reveal how photoexcitation increases their potency, and connect these catalysts to light-harvesting antennas. We understand the fundamental photophysics of high electrochemical potential generation for photocatalytic reactions; devise sophisticated new mechanisms to harness the energy of multiple photons and multielectron transfers to actuate redox states in Thrust B (Lead: Garry Rumbles). We elucidate photophysical mechanisms by which photosynthetic proteins use light harvesting as a distribution hub for excitation energy in Thrust C (Lead: Gabriela Schlau-Cohen) and design biohybrids that function as biocatalysts for high-energy chemistry. The molecular insights obtained from Thrusts A – C will be utilized to design and demonstrate photoinduced cross-coupling reactions using the combined energy of multiple photoexcitations to drive chemical reactions that are prohibitively disfavored at equilibrium in Thrust D (Lead: David MacMillan). In particular, we target bond activation by organometallic catalysts to produce chemical feedstocks and fuels.

In the past half-century, transition metal catalysis has arisen as a uniquely enabling platform for molecular construction. The development of *cross-coupling* technologies, wherein aryl or alkyl substrates can be forged together, have proven to be particularly powerful for the rapid construction of complex molecular frameworks from modular building blocks. Simply put, these reactions enable the attachment of two molecular units A and B to form the new molecule A–B, facilitated by organometallic reagents (Figure 2).

A common reactive precursor for cross-coupling technologies includes a carbon–halogen bond, because that bond is readily ‘activated’ (i.e. the functional group can be edited). However, production of this precursor requires energy-intensive synthesis and it generates hazardous waste. Chemical feedstocks should be readily transformable under current synthetic methods. For example, phenols and anilines are abundant, naturally occurring compounds that can be found in biomass feedstocks, but at present their functional groups (–OH or –NH_2), unlike halides, are too strongly attached to the phenol ring to be activated. The sophisticated photoinduced cross-coupling chemistry reactions needed are currently inconceivable using today’s leading-edge synthetic chemistry. These challenges are being addressed by BioLEC. For example, we hypothesize that we can employ phenols and anilines as aryl halide surrogates in cross-coupling reactions, with oxidative addition activating the traditionally unreactive C–O and C–N bonds, respectively. That will be enabled by working out how to escalate the chemical reactivity of organometallic catalysts by using a combination of photo-excitation and photoinduced bimolecular electron transfers to yield nonequilibrium valence states ‘on the fly’ during the reaction.

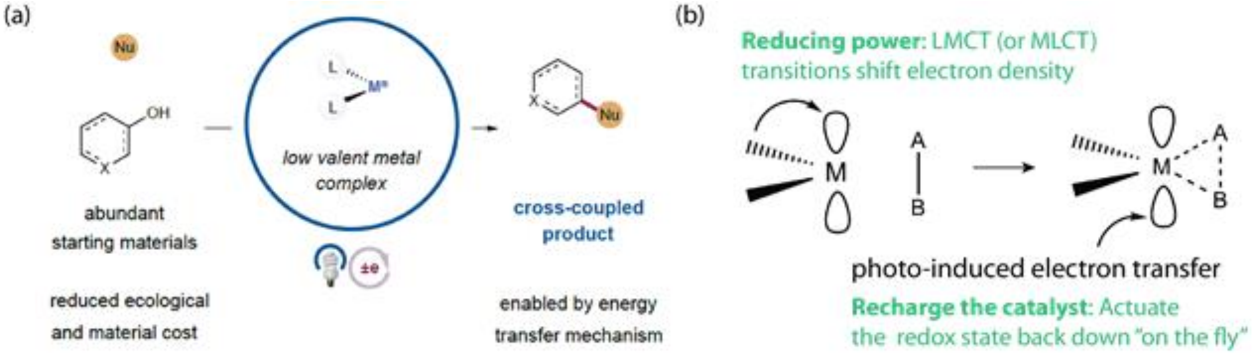

Figure 2. (a) Activation of inert bonds for cross-coupling. (b) BioLEC will step-change the limits of equilibrium redox potentials by adding or removing electrons “on the fly,” thereby driving reactions forward.

Bioinspired Light-Escalated Chemistry (BioLEC)	
Princeton University	Gregory Scholes (Director), David MacMillan (Scientific Director), Abigail Doyle, Todd Hyster, Robert Knowles, Barry Rand
Massachusetts Institute of Technology	Gabriela Schlau-Cohen (Associate Director)
North Carolina State University	Felix Castellano
Brookhaven National Laboratory	Matt Bird
National Renewable Energy Laboratory	Garry Rumbles
Arizona State University	Ana Moore, Thomas Moore
Michigan State University	James McCusker

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Photonics at Thermodynamic Limits (PTL)
EFRC Director: Jennifer Dionne
Lead Institution: Stanford University
Class: 2018 – 2022

Mission Statement: *To achieve photonic operations at thermodynamic limits by controlling the flow of photons, electrons, and phonons in atomically-architected materials, enabling entirely new energy conversion systems.*

Thermodynamic cycles enable optimized performance of nearly every energy conversion device that underpins advanced economies. While most thermodynamic cycles rely on a classical fluid, photons can also be used to drive thermodynamic cycles. The “Photonics at Thermodynamic Limits” (PTL) EFRC strives to achieve photonic operations at thermodynamic limits by controlling the flow of photons, electrons, and phonons in atomically-architected materials, enabling entirely new energy conversion systems. Such photon-based Carnot cycles offer remarkable opportunities for energy conversion, including all-optical energy-storage, optical refrigeration, optical rectification, power-generating windows that absorb light yet remain transparent, and beyond von-Neumann information architectures. Realizing photonic thermodynamic cycles requires new optical materials design, synthesis and characterization so that photonic operations – such as absorption, emission, and reflection – can be performed with the highest possible efficiency.

Objectives for 2018-2022

To design new photonic energy systems based on very high radiative efficiency, the PTL EFRC uses theory to guide experiments that are in turn validated by state-of-the-art characterization techniques. We will:

1. Design and develop atomically-precise and ‘beyond-ideal’ materials that perform photonic operations at thermodynamic limits (Research Group 1).
2. Develop transformative characterization methods to correlate structure-to-function with unprecedented spatial and temporal resolution (Research Group 2)
3. Investigate emergent physical phenomena and photonic thermodynamic cycles that arise when photonic processes approach the thermodynamic limits of photonic operations (Research Group 3).

Center Research Team and Scientific Organization

To achieve our mission, we have united leading researchers in layered and nanostructured materials synthesis, electromagnetic theory, first-principles quantum theory of materials, and advanced characterization of excited state phenomena. The Center is organized scientifically into three research groups (RGs) that address scientific themes related to photonics at thermodynamic limits, with each team spanning multiple institutional partners and designed to address our four-year scientific objectives.

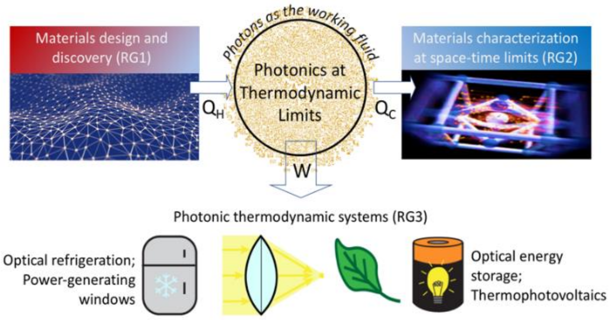

Figure 1: *Schematic overview of our “Photonics at Thermodynamic Limits” Energy Frontier Research Center.*

- **Research Group 1 – Materials Design and Discovery:** Using novel quantum-electrodynamic and first-principles calculations, the RG1 team is identifying conditions that maximize the useful work of a photonic system, which involves the energy, momenta, phase, and entropy of photons. In parallel, using state-of-the-art materials chemistry, we are developing nanoparticles and 2D materials with near-unity quantum yields, ultra-high-conductivity and reflectivity, and low-power nonlinearities.

- **Research Group 2 – Materials Characterization at Space-Time Limits:** The RG2 team is developing transformative characterization methods that include (1) novel in-operando characterization platforms based on optical microscopy within a transmission electron microscope (OTEM); (2) ultrafast stimulated emission depletion (STED) optical microscopy; (3) photothermal deflection spectroscopy; and (4) single particle ultrafast diffraction leveraging the unique facilities at SLAC National Accelerator Laboratory.
- **Research Group 3 – Photonic Thermodynamic Systems:** The RG3 team will investigate emergent physical phenomena and photonic thermodynamic cycles that arise when photonic processes approach the thermodynamic limits of photonic operations, including radiative cooling, all-optical energy storage, power-generating windows, and thermophotovoltaics.

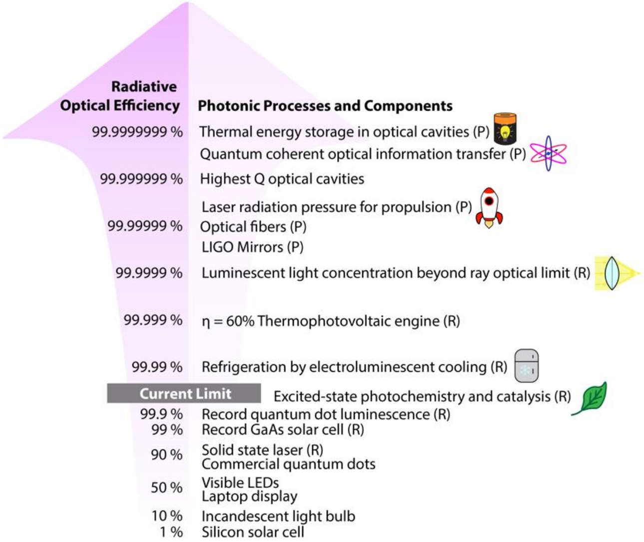

Figure 2: *Passive (P) and Active radiative (R) systems that become possible with increasingly high optical efficiency.*

Selected Accomplishments

Our long-term goal is to design photonic conversion systems for energy and information that operate at thermodynamic limits, and to share our research with technologists, policymakers and the public to maximize the societal impact of our EFRC. Selected accomplishments to date include:

- Achieving record conversion efficiency in a regenerative thermophotovoltaic system, incorporating photonic design to reuse low-energy photons (RG3).
- Realizing near-unity luminescence in core/shell quantum dots (RG1) and characterizing their quantum yield with high-precision using photothermal threshold quantum yield measurements (RG2).
- Designing novel heterostructures of two-dimensional transition metal dichalcogenides (RG1).
- Correlating optical and electron microscopy to identify the role of structure on quantum emission in two-dimensional materials (RG2).
- Developing a quantum theoretical description of electron/phonon and photon/phonon dynamics in nanoparticles and 2D materials (RG1).
- Utilizing ultrafast electron diffraction (UED) at SLAC to unravel nanocrystal structural dynamics (RG2).

Photonics at Thermodynamic Limits (PTL)	
Stanford	Jennifer Dionne (Director), Shanhui Fan (RG3 Leader), Tony Heinz (RG1 Leader), Mark Brongersma, Aaron Lindenberg, Alberto Salleo
UC Berkeley	Paul Alivisatos, Naomi Ginsberg (RG2 Leader), Eran Rabani, Eli Yablonovitch
Caltech	Harry Atwater (Deputy Director), Carrie Hofmann (Associate Director)
Harvard	Prineha Narang
UIUC	Ralph Nuzzo, Paul Braun (Adjunct Investigator)

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Center for Mechanistic Control of Water-Hydrocarbon-Rock Interactions in Unconventional and Tight Oil Formations (CMC-UF)

EFRC Director: Anthony R. Kovscek
Lead Institution: Stanford University
Class: 2018 – 2022

Mission Statement*: To seek fundamental mechanistic understanding to achieve control over the various non-equilibrium chemical and physical processes occurring in shale that increases hydrocarbon production while decreasing the amount of produced water, contaminants, and the number of wells drilled.*

This EFRC features a tightly integrated fundamental research program combining experimental, theoretical, and numerical science. The plan is comprehensive and interdisciplinary. Importantly, our five overarching research themes foster a fundamental knowledge base from which we learn how to characterize and control single and multiphase reactive transport in shale that is far from equilibrium. We have crafted our plan to unravel heterogeneity and fluid interactions with shale mineral/organic-matter interfaces within the context of a natural extremely disordered system as a function of stress and transport. We exploit and create new imaging and image reconstruction capabilities to explore the largely inaccessible interior of the shale matrix. This is in itself a 'frontier' research focus area. Characterization of nanoporous disordered media before, during, and after reaction is coupled with modeling at length and time scales of interest using advanced models, algorithms, upscaling methods, and computing. The figure below overviews the interplay of length scales, characterization, and integrative modeling activities. It shows the 10 orders of magnitude in length scale this investigation spans. Scale translation is a unifying activity within the research plan as indicated by the progression of scales. Machine learning and data analytics feature throughout the center as important tools for improving physical insight.

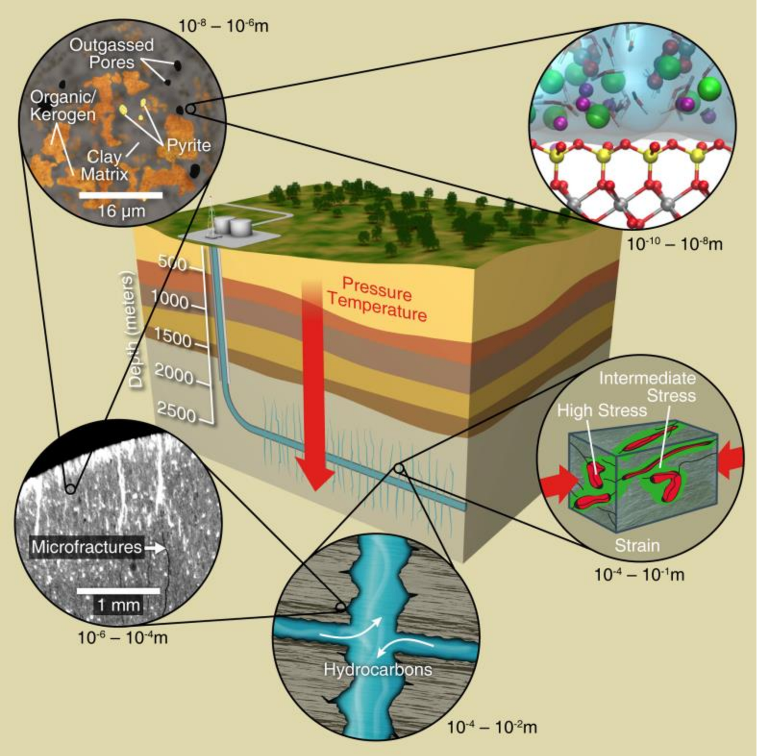

Figure. Overlapping length scales of shale features of interest to this EFRC. *From the upper right*: water/clay interactions in a 10 nm wide slit pore where water is shaded blue; nanoCT image of shale structure illustrating minerals, kerogen, and outgassed pores; microfractures filled with barite scale; matrix-to-fracture mass transfer; zones of enhanced ductility in heterogeneous shale.

To control materials and processes we must understand mechanisms and use this fundamental knowledge to predict how processes function and evolve with time in diverse chemical and stress state environments. The fundamental science understanding of the coupled chemical and physical processes involving water, hydrocarbons, and substitute fracturing fluids at interfaces in nanoporous media is immature. This Center uses a bottom-up, multiscale, multiphysics, and multidisciplinary approach to investigate disordered nanoporous media incorporating and integrating experiments, structural and chemical characterization before and after reactions, and theory. Scale translation of experimental and model results serves as the centerpiece of our integrative activities.

The center is organized around five cross-disciplinary science goals.

1. Develop and exploit advanced multiscale imaging capabilities using x-ray, electron microscopy, and nuclear magnetic resonance techniques to characterize and analyze the fabric of shale at nm to cm scales
2. Elucidate the coupled phase behavior, geomechanical, and transport mechanisms of single and multiphase flow through shale using length-scale appropriate experiments and models to understand the controls on flow and transport of water and hydrocarbons.
3. Measure, characterize, and model aqueous fluid interactions at shale mineral interfaces and the influence of water composition on matrix, microfracture, and fracture fluid transfer and transport.
4. Characterize the mechanisms of viscoplasticity and ductility of shale when exposed to alternate hydraulic fracturing fluids such as CO_2, N_2, and aqueous foams of CO_2 and N_2.
5. Enable translation of physical and chemical mechanisms to assess their influence at macroscopic length and time scales using advanced algorithms and modeling that take advantage of emerging high-performance computing with heterogeneous processors and complex memory hierarchies.

Center for Mechanistic Control of Water-Hydrocarbon-Rock Interactions in Unconventional and Tight Oil Formations (CMC-UF)	
Stanford University	Anthony R. Kovscek, Ilenia Battiato, Sally Benson, Matthias Ihme, Hamdi Tchelepi, Mark Zoback
University of Wyoming	Vladimir Alvarado, Saman Aryana, Teresa Lehmann
University of Southern California	Kristian Jessen, Theo T. Tsotsis
University of Illinois	Jennifer Druhan
SLAC National Accelerator Laboratory	John Bargar, Gordon Brown

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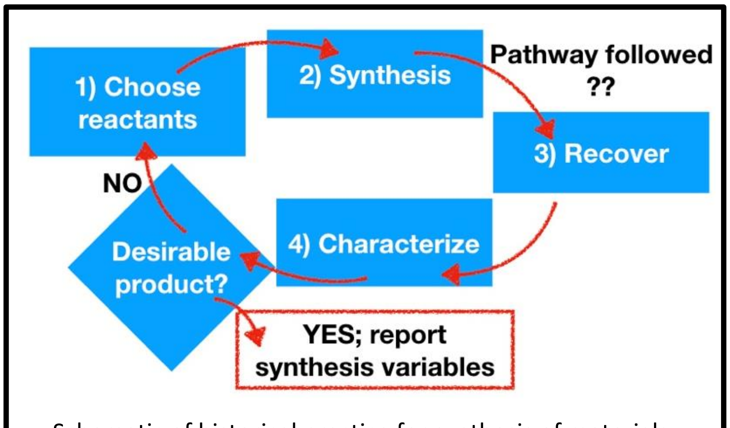
A Next Generation Synthesis Center (GENESIS)

EFRC Director: John B. Parise
Lead Institution: Stony Brook University
Class: 2018 – 2022

Mission Statement*: To develop a new paradigm for synthesis that accelerates the discovery of functional materials by integrating advanced in situ diagnostics and data science tools to interrogate, predict, and control the pathways that govern synthesis and lead to new materials.*

The “cook-and-look” technique remains the mainstay of materials research and development. Researchers seal chemical reactants in a vessel, “cook” to initiate a reaction and after some time “look” at the recovered products to determine if they are in a form required to be useful.

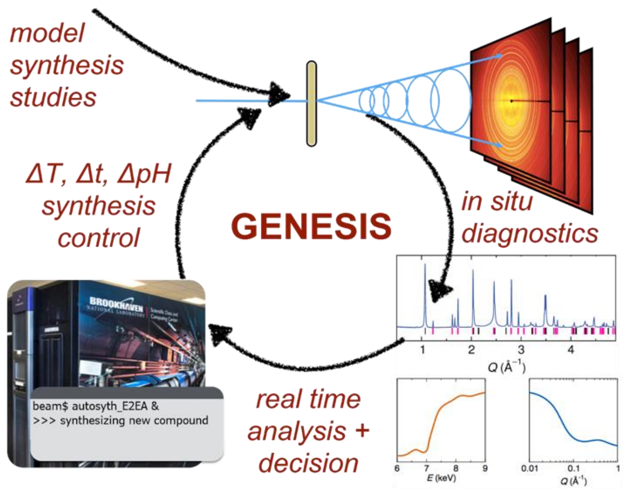


Schematic of historical practice for synthesis of materials.

The formidable task of repeated synthesis-recovery-characterization can be accelerated using modern in situ techniques that allow us to “look inside” the reaction vessel. The paths taken by a reaction from starting materials to final functional product are opaque to researchers without the use of in situ techniques. However, the data, such as those collected using beams capable of penetrating reaction vessels at DOE X-ray and neutron User Facilities, are mostly analyzed off-line. During reactions transient species form, grow and transform to other species. These processes are critical to the final product and all are invisible without the ability to follow them in situ *and* in real time. Observation of the reaction pathway and real time analysis of data reveals the fundamental mechanisms that result in the final product, at the molecular level and step by step. The science of synthesis lies in not only mapping the reaction pathway but also in understanding at the atomic level the underlying operational mechanisms that occur all the way along the pathway; determining, at the speed of the reaction, which atoms are doing what. That requires the development of computational approaches that identify what phases are forming when. In order to speed the development of transformational materials the challenge of tracking the evolution of phases along the reaction pathways must be met. Importantly, knowing the details of the reaction pathway allows us to steer the reaction in new directions, towards novel functional materials not yet realized.

Beyond implementation of computational approaches for the real time analysis of reaction pathways can we develop a data-driven approach to initiate desired synthesis pathways on-demand? Because theory struggles to describe the non-equilibrium diffusional processes governing real syntheses due to the incredibly complex, dynamically-evolving, defect-driven multi-dimensional parameter space associated with the synthesis mechanism of real materials, a data science approach is going to be more fruitful. Can we build a framework that moves us forward from think-cook-look-repeat strategies to machine-suggested synthesis pathways, in a manner analogous to now-mature computational structure- and properties-prediction methods?

The tremendous amounts of minable data on reaction pathways produced by in situ experiments, not only by GENESIS but by the community of scientist worldwide, can be mined using Natural Language (NL) searches. GENESIS will capture all synthesis parameters, including temperature history, process gas composition and flow rates, concentrations of all liquid species, and the composition, into structured-data databases. These data will become features for supervised learning algorithms in NL that will enable us to use an efficient, data-intensive approach and find elements for new pathways in publications and databases: the structure, defects, and morphological parameters (size, crystallinity) of solid phases thus discovered will also be captured.



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beam$ autosyth_E2EA &
>>> synthesizing new compound
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GENESIS workflow illustrating that in situ probes are analyzed in real time to yield controlled, next-generation synthesis.

By this approach, GENESIS builds the tools and understanding to discover new materials more rapidly. In the end, to be useful, a material must be made, and control over synthesis pathways and products is a critical requirement for materials design, without which concepts developed in computers cannot be brought into reality. Computational approaches can now identify a library of potential materials; however, synthesis and physical realization of these new predicted materials remains a critical limit. The GENESIS approach eliminates this bottleneck by exploiting advanced real time diagnostics coupled with data science tools and, thereby, accelerate the synthesis of new materials. The acquisition of large databases of high value synthetic data will be accelerated if the input parameter space is searched in a "smart" way, rather than by brute force. Indeed, the high dimensionality of the input parameter space makes this essential. A longer-term goal of GENESIS is to develop adaptive experiment controls-- the ability of the control software to "steer" reactions in real time based on the results of just run experiments. These tools will be made available to the wider community so that different groups can carry out similar investigations using the hardware and computational infrastructure developed in GENESIS.

A Next Generation Synthesis Center (GENESIS)	
Stony Brook University	John Parise (Director), Karena Chapman (Associate Director), Peter Chupas, Peter Khalifah, Brian Phillips
Brookhaven National Laboratory	Line Pouchard, Eric Dooryhee
Columbia University	Simon Billinge
Colorado State University	James Neilson
Farmingdale State College	Jack Simonson
Lawrence Berkeley National Laboratory	Kristin Persson
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Center for Mesoscale Transport Properties (m2m/t)
EFRC Director: Esther S. Takeuchi
Lead Institution: Stony Brook University
Class: 2014 – 2022

Mission Statement*: To build the scientific knowledge to enable creation of scalable electrochemical energy storage systems with high energy, power, and long life.*

The research conducted under this award (m2m/t, molecular to mesoscale over time) will build the necessary insights to move beyond the flat electrodes in widespread use today to achieve the mission. In particular, it is imperative to understand and address transport limitations of multiple electrode architectures in electrochemical systems and their evolution over time. In order to achieve scalable electrochemical energy storage systems, facile ion transport and electron transfer are essential. The research conducted by the *m2m/t* Center will overcome current barriers resulting from existing knowledge gaps. The Center will pursue three Science Goals (SG) where each will be achieved through specific Research Objectives. The *Science Goals* (SG) are listed below where active material design is pursued in SG1 to facilitate transport and cycle life. SG2 addresses interface design and stability, to gain fundamental understanding of key considerations for multiple electrode designs. SG3 explores electrode architectures enabling ion access from the electrolyte to minimize transport losses in reaching all active material particles. The Science Goals represent interactive initiatives where SG1 and SG2 will ultimately enable the full realization of SG3 addressing the Center mission.

SG1. Design and create innovative multifunctional materials that synergistically integrate the multiple functions provided by individual components in battery electrodes.

SG2. Understand and control kinetically driven interfacial phenomena by deliberate design and manipulation of dynamic interfaces.

SG3. Gain fundamental understanding of transport properties in order to rationally design three-dimensional architectures.

Electrochemical function will be investigated with state-of- the-art tools, including *in-situ* and *operando* methods, where spatial and temporal resolution will be utilized to understand transport and transport limitations. Our focused and integrated effort with expertise in materials, theory, modeling and experimentation will deliver meaningful insights to benefit both current and next generation battery systems.

Approach: The *m2m/t* Center will achieve the goals via: 1) Synergistic interaction of experts in materials, characterization, theory, modeling, and electrochemical systems analysis; 2) Deployment of revolutionary analytical tools including electron microscopy and photon science with unprecedented resolution as well as real-time data acquisition capabilities; and 3) Investigation unified by innovative material concepts and versatile electrode constructs.

Under Science Goal 1 the research will probe several critical science questions. Can multifunctional materials be successfully designed and synthesized? How do electrical conductivities compare to the native oxides? Will integrating multiple functionalities into these materials result in enhanced electrochemical transport properties in electrochemical energy storage devices? Can electrochemical cycle stability be enhanced through purposeful material design and manipulation?

Under Science Goal 2 the research will pursue the following questions. Will deliberately designed and synthesized model electrode structures enable interface, properties, and time-dependent evolution to be interrogated using high signal-to-noise bulk measurements? Will advanced analytical approaches allow characterization of dynamic interfaces of electrochemical energy storage systems? Using insights gained, can interfacial phases with specific chemical composition be created for use in model electrochemical systems suitable for quantitative comparisons and refinement of multiscale models?

The activities under Science Goal 3 will investigate the following questions. Can targeted design of 3D electrode architectures overcome ion transport limitations to enable high power and high capacity scalable designs? Will exploitation of state-of-the-art in-situ and operando methodologies to characterize materials properties and electrode dynamics under realistic operating conditions provide sufficient spatio-temporal resolution to develop continuum models? Will integration of existing and new computational techniques adequately predict temporal evolution of spatially distributed properties to advance next generation energy storage?

The Center is pursuing these critical issues to move beyond conventional electrode architectures based on flat 2D layers where active materials interact through short distances and over large lateral areas. The proposed research tackles fundamental limitations of incomplete active materials reaction and sluggish transport kinetics through considering the multiple length scales critical to electrode design and function, **Figure 1**. The Center research will advance function-oriented electrode design *via* integrated experimental and theoretical approaches, including rational material design, structural engineering through novel synthesis, interface stabilization and advanced *ex-situ*, *in-situ*, and *operando* characterization tools linked with comprehensive theory and continuum modeling. This integrated effort will empower the community to move from serendipitous trial and error to rational design, and from compartmentalized knowledge to integrated understanding.

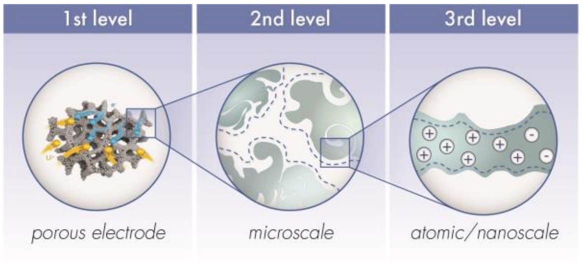

Figure 1. Multiscale transport in electrode architectures.

Center for Mesoscale Transport Properties (m2m/t)	
Stony Brook University	Esther Takeuchi (Director), Amy Marschilok (Center Operations Officer) Kenneth Takeuchi, Stanislaus Wong, Karen Chen-Wiegart
Brookhaven National Laboratory	David Bock, Ping Liu, Feng Wang, Lei Wang, Yimei Zhu
Columbia University	Alan West
Cornell University	Lynden Archer
Drexel University	Ekaterina Pomerantseva
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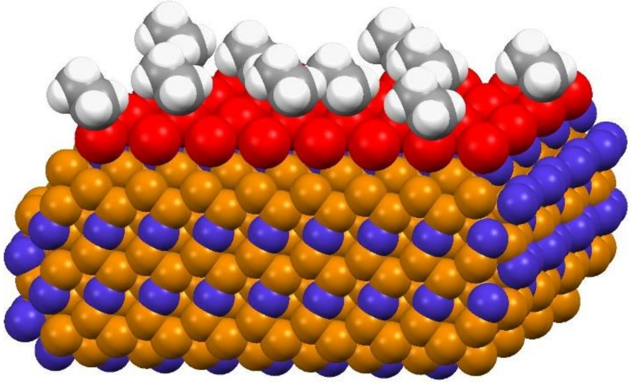
Center for Complex Materials from First Principles (CCM)
EFRC Director: John P. Perdew
Lead Institution: Temple University
Class: 2014 – 2020

Mission Statement*: To develop, test, apply, and experimentally validate improved methods of electronic structure calculation for both simple and complex materials (including quantum materials).*

BACKGROUND: EFRC CCM is an extension of the 2014-2018 *Center for the Computational Design of Functional Layered Materials (CCDM)*, which demonstrated that computationally-efficient first-principles density functionals can work well not only for simple materials but even for many complex ones.

CCM brings together world-class scientists from different disciplines to tackle problems in the field of complex materials in new ways. To this end, CCM integrates state-of-the-art **Theory (Thrust 1)** with **Modeling and Computation (Thrust 2)** and **Experiment (Thrust 3)** to enable materials property predictions that are linked to **Experiment (Thrust 3)**, to accelerate the rate of progress on complex (including quantum) materials. Integration of the three thrusts is further enhanced by cross-cutting activities of **Forum A: Properties** and **Forum B: Applications.**

CCM will develop, test, apply, and experimentally validate improved methods of electronic structure calculation for both simple and complex materials. More accurate but still computationally efficient methods will be useful to many scientists. In particular, these methods will increase the reliability and expand the range of high-throughput computational searches for materials with desired properties. Because they will be developed from first principles, the new methods will be reliable and widely predictive. Planned tests and applications include the layered and two-dimensional materials, the cuprate high-temperature superconducting materials, water at interfaces, catalysis, and materials synthesis.



A material is defined by its chemical composition and structure. Complex materials are those in which small and difficult-to-predict energy differences per atom can imply large qualitative changes in other properties, leading in many cases to rich phase diagrams (and often to enlarged unit cells). Easy switching between phases can be exploited for some practical applications. For other applications, the relative stability of simple materials is advantageous. There is of course a continuum from simple to complex materials. Predictive theoretical methods should apply to both kinds of materials (and to surfaces, clusters, and molecules), and thus must be based upon the first principles of quantum mechanics. These methods should range from the more highly-accurate ones that can provide benchmarks to the more computationally-efficient ones that permit wider searches over the enormous space of possible materials. Complex materials include the quantum (e.g., low-dimensional and cuprate) materials, but also more familiar ones like ice (or other molecular crystals) and liquid water at interfaces. CCM will develop some benchmark methods, such as a coupled cluster or random phase approximation, but will mainly focus on the more computationally-efficient density functional methods. Better density functionals for complex materials will imply better theoretical descriptions of interfaces and catalysis, and new targets for materials synthesis.

Density functional theory can in principle determine the exact ground-state properties for interacting electrons subject to static scalar external (possibly spin-dependent) potentials, via solution of self-consistent one-electron equations. Accessible properties include electron spin densities, total energy, fundamental energy gap in a generalized Kohn-Sham implementation, and forces on nuclei. Within the adiabatic and classical-nuclei approximations, this theory also determines vibrational properties of nuclei, and serves as a foundation for *ab initio* molecular dynamics. Many-body effects are taken into account, not through a computationally-expensive correlated wavefunction, but through a density functional for the exchange-correlation energy, whose functional derivative is an exchange-correlation potential. This functional is “Nature’s Glue”, providing most of the binding between atoms in a material or molecule. The exact functional is well-defined but un-computable in practice. Simple approximations to it however provide a practical and often usefully accurate way to predict binding energies, equilibrium geometries, equations of state, phase transitions, vibrational frequencies, etc., for many materials or molecules.

Until recently, accurate non-empirical density functional calculations were only possible for simple materials. Work done in CCDM showed that density functional approximations constructed to share many properties of the exact functional (exact constraints) can be accurate even for some complex materials. CCM will extend this work by making better functionals, then validating and applying them more widely.

Center for Complex Materials from First Principles (CCM)	
Temple University	John P. Perdew (Director), Eric Borguet (Deputy Director, Experiment Thrust Leader), Maria Iavarone (Deputy Director), Michael L. Klein, Adrienn Ruzsinszky, Daniel R. Strongin (Applications Forum Leader), Xifan Wu, Xiaoxing Xi, Qimin Yan, Michael J. Zdilla
Brookhaven National Laboratory	Yimei Zhu
Drexel University	Goran Karapetrov
Duke University	Weitao Yang
Northeastern University	Arun Bansil (Properties Forum Leader)
Rice University	Gustavo E. Scuseria (Theory Thrust Leader)
Tulane University	Jianwei Sun
University of Pennsylvania	David J. Srolovitz (Modeling and Computation Thrust Leader)

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Center for Gas Separations (CGS)
EFRC Director: Jeffrey R. Long
Lead Institution: University of California, Berkeley
Class: 2009 – 2020

Mission Statement*: To develop new materials and membranes that enable the energy-efficient separation of gas mixtures, as required in the clean use of fossil fuels and in reducing CO_2 emissions from industry.*

Separation processes have been estimated to account for 10–15% of our total energy consumption. As the global population grows and our reliance on fossil fuels continues in the near term, this percentage is anticipated to increase significantly, both as raw material demand increases and as we make essential investment in large-scale carbon capture and sequestration technologies. Reducing the total energy costs associated with various separations in industry and powder generation would contribute significantly toward lowering our overall energy usage. The aim of the Center for Gas Separations (CGS) is to develop new porous materials that enable energy-efficient gas separations, based on unprecedented molecule-specific interactions, together with new and powerful characterization methods that provide a fundamental understanding of the properties and performance of such materials.

The challenge is to remove the fundamental scientific barriers that currently prohibit the energy-efficient separation of gases essential to the development of clean energy technologies. This challenge is significant, as the chemical and physical differences between molecules in gas mixtures of interest are often small, and therefore it is necessary, through the use of nanoscience and synthetic chemistry, to engineer unprecedented molecular-level control in adsorbate–adsorbent interactions and thereby tailor-make 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:

- *Materials Synthesis*: The synthesis of new gas-permeable materials with control over the molecular functionalities is essential to enable preferential adsorption of gas molecules. Our focus here is 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 is necessary both before and after exposure to gas samples in order to probe interaction mechanisms and further tailor material properties. We develop novel characterization methods that enable us to accurately assess the selectivity, kinetics, and thermodynamics of gas adsorbate binding, both to demonstrate efficacy and test computational models.
- *Computational Separations*: A strong computational component to the research is essential for understanding chemical interactions at a molecular level, as well as for guiding the synthetic efforts toward materials exhibiting high specificity for a given adsorbate and tunable interaction energies.

We are developing exceptional new materials for a host of 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 O_2 from air, the separation of N_2 from natural gas, and the separation of industrially relevant hydrocarbon mixtures.

Center for Gas Separations (CGS)	
University of California, Berkeley	Jeffrey Long (Director), Jeffrey Neaton, Jeffrey Reimer (Deputy Director), Berend Smit, Ting Xu, Omar Yaghi
Lawrence Berkeley National Laboratory	Walter Drisdell, Maciej Haranczyk, Brett Helms, David Prendergast, Simon Teat, Steve Whitelam
Texas A&M University	Hong-Cai Zhou
University of Minnesota	Laura Gagliardi, Michael Tsapatsis
National Energy Technology Laboratory	David Hopkinson
National Institute of Standards and Technology	Craig Brown
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Synthetic Control Across Length-scales for Advancing Rechargeables (SCALAR)
EFRC Director: Sarah Tolbert
Lead Institution: University of California, Los Angeles
Class: 2018 – 2022

Mission Statement*: To use the power of synthetic materials chemistry to design materials, interfaces, and architectures that help solve long-standing problems in electrochemical energy storage.*

Lithium-ion batteries have dominated rechargeable electrical energy storage for over two decades. Fundamentally, however, materials for lithium-ion batteries have changed little during this time, even though both their applications and desired performance have expanded tremendously. The scientific mission of the SCALAR EFRC is based on leveraging the power of modern materials synthesis and characterization to create and understand, at a fundamental level, a new generation of battery materials that can overcome many of the limitations intrinsic to the archetypal intercalation hosts in use today. Within the SCALAR EFRC, the challenge we face is to rethink the chemistry associated with secondary ion batteries to dramatically expand the range of materials and chemistries that can be employed, to increase stability by controlling transport, and to control architectures and interfaces to enable the use of very high capacity materials.

The overarching center goal is to combine cutting edge synthetic methodologies with a multi-length scale view of energy storage to make non-incremental improvement in battery materials. From a broad perspective, our program seeks to:

1. Take a holistic approach to the design of new functional materials that bridges the atomistic, nanometer, and macro length-scales in the quest to improve battery performance
2. Leverage molecular and solid-state synthetic methods, combined with solution phase self-assembly, to create new electrode materials that increase capacity, reduce losses, and improve reversibility in rechargeable batteries

To address its mission, the SCALAR EFRC has defined five scientific objectives that represent particularly promising routes for achieving its goals. As a center, we aim to:

1. Synthesize new electrode materials that increase charge storage capacity by integrating the electrochemistry of anions and cations into a single material.
2. Reduce resistive losses in electrodes using doping to create conductive electrode materials and conductive binders and scaffolds to increase conductivity in composite electrodes.
3. Create nanoscale architectures and interfaces that improve reversibility by stabilizing electrodes against chemical degradation, facilitating desired reactions, and accommodating the morphological changes that occur upon cycling.
4. Design systems to optimize charge transfer processes across length-scales, from the atomic, to the particle, and finally to the electrode level.
5. Integrate molecular, solid-state, and self-assembly methods to create structures that achieve nanoscale control and atomic-scale precision in macroscopic architectures.

The center is organized in to three thrusts. Thrust 1, aimed at objective 1, uses a combination of anion cluster chemistry, anion intercalation chemistry, and anion redox in extended inorganic solids to integrate anion redox with cation redox in new materials. Thrust 2 attacks the problem of resistive losses outlined in objective 2 in two very different ways – first by exploring the use of conjugated polymers as binders with increased electrical and ionic conductivity, and second by exploring highly covalent and shear structures with enhanced ionic and electrical conductivity. These efforts are coupled with electrochemical calorimetry to directly measure thermal loss within our systems. Finally, Thrust 3 aims to solve the challenge of objective 3 by using inorganic surface coatings for improved stability and reversibility, using flexible nanoporous architectures that can mitigate and even compensate for large volume changes during cycling, and using catalysts that can favor desired electrochemical reactions and mitigate unfavorable ones.

Thrust level objectives (1-3) are combined with two cross-cutting objectives (4-5) focused on utilizing a diverse range of synthetic methodologies and a multi-length scale view of battery materials to design new materials that can be optimally integrated into battery electrodes. In carrying out the research mission, materials synthesis and design considerations are combined with cutting edge materials characterization and high level predictive modeling to create true synergy in materials design, atomistic materials synthesis, mesoscale structural control, and materials characterization across length-scales. By taking a holistic view of energy storage, the SCALAR EFRC will expand the understanding of battery materials and have a long term impact on how we think about the kinds of new materials and reactions that can be utilized for rechargeable electrical energy storage.

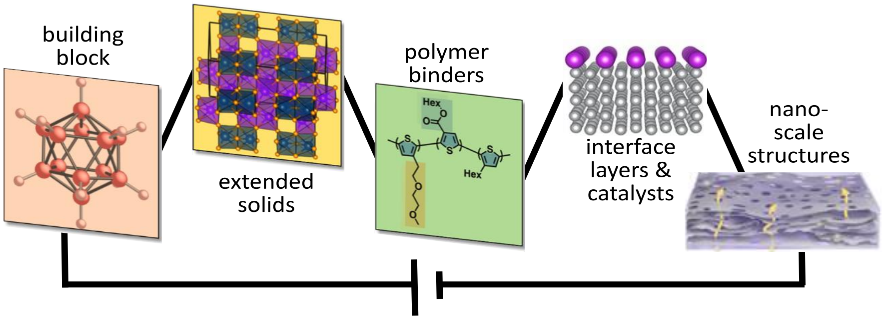

Figure 1: The SCALAR EFRC aims to rethink battery materials, starting from the level of building blocks and then moving to new motifs for extended solids. New materials are integrated with multifunctional polymer binders, designer interfaces, and controlled nanoscale architecture.

Beyond scientific synergy, the SCALAR EFRC takes advantage of the human synergy of the Southern California regional area, which houses a large number of world class research universities. Five of these universities, along with one California based national lab, have joined together to make the SCALAR center a regional hub for battery research that optimally leverages both the proximity and complementary facilities of the participating partner institutions.

Synthetic Control Across Length-scales for Advancing Rechargeables (SCALAR)	
University of California, Los Angeles	Sarah Tolbert (Director), Bruce Dunn (Deputy Director), Xuan Duan (Thrust 3 Lead), Laurent Pilon, Philip Sautet, Alexander Spokoyny
University of California, Santa Barbara	Ram Seshadri (Thrust 2 Lead), Bradley Chmelka, Rachel Segalman, Anton Van der Ven
University of Southern California	Brent Melot (Thrust 1 Lead), Sri Narayan, Barry Thompson
Caltech	Tom Miller, Kimberly See
Stanford Synchrotron Radiation Lightsource	Joanna Nelson Weker
University of California, San Diego	Jian Luo

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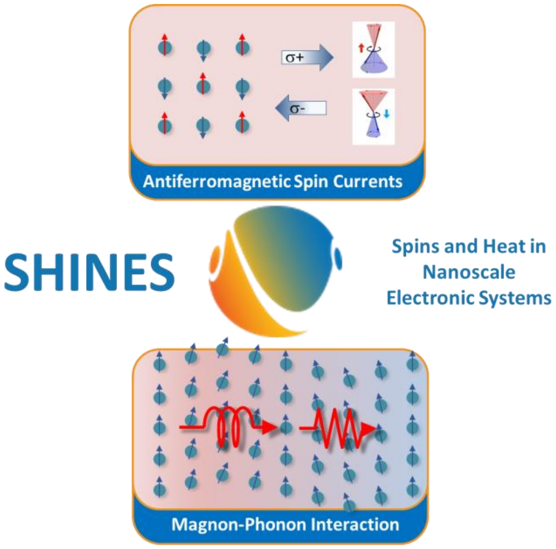
Spins and Heat in Nanoscale Electronic Systems (SHINES)

EFRC Director: Jing Shi
Lead Institution: University of California, Riverside
Class: 2014– 2020

Mission Statement: *To control interactions involving spins and lattice to achieve high energy efficiency in nanoscale electronic devices.*

Based on the progress made in the last four years, the renewed EFRC will focus on the understanding and control of fundamental interactions involving spins and lattice in magnetic thin films and heterostructures with the goal of increasing energy efficiency in nanoscale electronic and spintronic devices. Microscopic interactions such as spin-spin exchange interaction, spin-orbit coupling, and magnon-phonon interaction strongly affect or determine macroscopic properties of magnetic materials and spintronic devices such as magnetic anisotropy, spin-charge conversion efficiency, energy dissipation, etc. A research frontier in spintronics is discovering how to control these important interactions on the atomic scale by designing and synthesizing novel thin film materials and heterostructures. A great deal of progress has been made in SHINES since 2014 in advancing the spintronics frontier. For example, SHINES research has demonstrated that strong exchange interaction at the interface of magnetic and non-magnetic materials (e.g., magnetic insulators and topological insulators) can be created and manipulated through proximity coupling arising from electron wavefunction overlap between the two materials. The artificially created exchange interaction can then have profound consequences in the physical behaviors of the materials such as formation of chiral edge states, emergence of Majorana fermions, strong modification of spin relaxation and damping, etc. In the following two years, SHINES will continue exploring new materials (e.g., antiferromagnetic/topological insulator heterostructures) and new phenomena (e.g., exchange modulated ultrafast spin dynamics) that will lead to new functionalities and high energy efficiency in nanoscale electronic and spintronic devices.



The two-year objectives include discovering or demonstrating new spintronic phenomena in ferromagnetic and antiferromagnetic heterostructures, developing an in-depth understanding of these phenomena, controlling microscopic interactions via material growth and application of external stimuli such as electric and magnetic fields and therefore the resulting magnetic properties such as magnetic anisotropy, magnetic transition temperature, and damping. A special emphasis will be placed on the antiferromagnetic spintronics, an emerging research area that several SHINES investigators pioneered. The experimental techniques to be employed for this team research include molecular beam epitaxy and pulsed laser deposition for high-quality thin film and heterostructure growth, structural characterization using X-ray diffraction and neutron scattering, magnetic domain imaging, non-local transport, THz spectroscopy, magnetic resonance, spin pumping, spin Seebeck effect measurements, Raman and Brillouin light scattering, etc. The experimental investigations will be complemented with theoretical and computational work within SHINES through close collaboration.

Spins and Heat in Nanoscale Electronic Systems (SHINES)	
University of California, Riverside	Jing Shi (Director), Alex Balandin, Roger Lake, Igor Barsukov
University of California, Los Angeles	Kang L. Wang
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Quantum-Materials for Energy Efficient Neuromorphic- Computing (Q-MEEN-C)

EFRC Director: Prof. Ivan K. Schuller
Lead Institution: University of California, San Diego
Class: 2018 – 2022

Mission Statement: *To lay down the quantum-materials-based foundation for the development of an energy-efficient, fault-tolerant computer that is inspired and works like the brain (“neuromorphic”).*

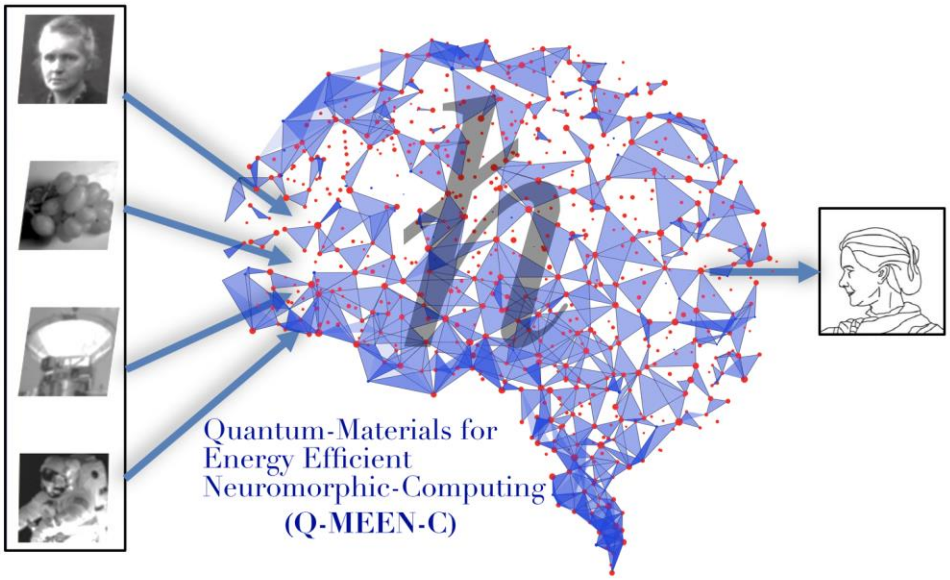

Fig. 1: Q-MEEN-C will develop the next-generation energy-efficient quantum-materials-based platforms which will be the basis of neuromorphic computing. In this example of neuromorphic computing, the image of a famous scientist is readily distilled from multiple disparate, complex inputs.

Energy-efficient neuromorphic computing offers a potentially disruptive technological capability to process complex inputs and produce elegantly simple, useful outputs as illustrated in Fig. 1. The breakaway from the conventional technology Turing-von Neumann paradigm requires the development of new types of bio-inspired (“neuromorphic”) devices with functionalities like artificial synapses, neurons, axons, and dendrites that can be used to construct machines with artificial intelligence capabilities. We aim to address these critical issues on two different platforms based on *Charge*- and *Spin*-related phenomena in quantum materials.

Technical Implementation: The *Charge*-based approach relies mainly on nanostructured quantum materials, which exhibit spectacular, unexpected changes in their electrical properties when subject to temperature, electric, and magnetic fields, strains, and light. Essential components of neuromorphic information processors will include artificial neurons, synapses, axons, and dendrites to mimic biological entities for transmission and processing of signals. These types of components will be realized in simple transition metal oxides or complex strongly correlated oxides, which exhibit metal-insulator transitions (MIT). In this fashion, we will aim to emulate typical neuronal activities, such as leaky-integrate-fire behavior, symmetric and asymmetric synaptic plasticity, and self-sustained oscillations. Eventually, several of these will be incorporated into complex circuits and architecture. Specific goals of these part of the EFRC are: 1) Identifying material platforms that can be controlled by non-thermal means such as electric fields, currents, strain, and light, 2) understanding ionic transport under highly non-equilibrium conditions, and 3) determining the ultimate limitations of MIT in these materials platforms.

The *Spin*-based approach relies on the implementation of neurons and synapses using heterostructured spin-torque oscillators, based on quantum-materials phenomena such as spin-orbit scattering. Neurons are emulated with magnetic oscillators, and the coupling between them mimics synapses. A charge current sent through these oscillators is converted into a spin current that drives magnetization precession of nanoscale structures in a sustained and coherent way at room temperature. The underlying magnetization dynamics are highly non-linear, and tunable in phase, amplitude and frequency. Therefore, spintronic nano-oscillators can strongly modify their dynamics in response to small incoming signals resulting in outstanding phase locking and mutual synchronization properties. This is key for neural networks, where neurons should respond to external inputs, but also to signals coming from other neurons through synapses. Specific goals of these part of the EFRC are: 1) Design materials for efficient operation of oscillators, 2) Develop approaches that are best suited for reconfigurable coupling between oscillators and 3) Engineer large-scale oscillator networks that generate complex dynamics.

The scientific thrusts will be interconnected with cross-cutting methods spanning novel materials design, state-of-the-art characterization, modern theory and computation (Fig. 2). A key strength of Q-MEEN-C combines these approaches to understand and harness 'designer' quantum material-based heterostructures with useful behavior for developing artificial, energy-efficient neurons, synapses, and axons. This will be achieved through multimodal imaging, spectroscopy, and diffraction techniques, coordinating with sophisticated modeling and simulation tools.

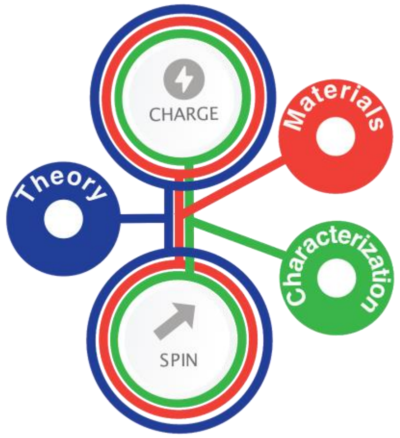

Fig. 2: Integration of our scientific thrusts with cross-cutting tools.

Byproducts: The research on quantum materials to be performed will produce as "byproducts" important basic research results relevant to electromigration, materials-property prediction, novel tools, and new materials and functionalities. From the technical point of view, the coordinated application of a large battery of synthesis, characterization, and theoretical tools makes this a comprehensive center that could not be successful without such an interdisciplinary, collaborative effort.

Quantum-Materials for Energy Efficient Neuromorphic- Computing (Q-MEEN-C)	
University of California, San Diego	Ivan Schuller (Director), Oleg Shpyrko (Associate Director), Alex Frano (Assistant Director), Robert Dynes, Y. Fainman, Eric Fullerton, Vtaliy Lomakin, Shyue Ping Ong
University of California, Davis	Yayoi Takamura
University of California, Santa Barbara	Jonathan Schuller
University of Chicago	Giulia Galli
Brookhaven National Laboratory	Y. Zhu
Purdue University	Shriram Ramanathan
New York University	Andrew Kent
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Catalysis Center for Energy Innovation (CCEI)
EFRC Director: Dionisios G. Vlachos
Lead Institution: University of Delaware
Class: 2009 – 2022

***Mission Statement**: To advance the catalysis science of complex systems with a focus on thermocatalytic transformation of lignocellulosic (non-food-based) biomass into chemicals and transportation fuels.*

CCEI has established itself as a transformative catalysis center with a mission to advance fundamental catalysis science in complex systems. We envision innovations toward feedstock diversification for co-processing of biomass with shale gas derivatives. To that end, we have established three new downstream research thrusts for the synthesis of: (1) *dienes* via dehydra-decyclization of oxacyclopentanes (saturated furans); (2) *surfactants* via acylation and aldol condensation of furan with fatty or shorter acids; and (3) *lubricants* via hydroxyalkylation/alkylation chemistry of furans with aldehydes and subsequent decyclization-hydrodeoxygenation of the intermediates. Major chemistry threads that emerge are: (1) C-C coupling chemistry to enable coupling of lignocellulose building blocks—among themselves or with shale gas derivatives; and (2) tandem furan ring opening and oxygen removal—as well as possible hydrogenation—to enable the design and synthesis of targeted functional molecules.

For the synthesis of dienes (e.g., butadiene)—important high-volume feedstock for polymers—we envision dehydra-decyclization of sugar-derived, oxacyclopentanes. To that end, we shall extend the use of *P-zeosils*, a new class of catalysts discovered by CCEI which consist of phosphoric acid supported on inert, all-silica zeolites. P-zeosils are Brønsted acids distinct from typical aluminosilicate zeolites, with P-speciation dynamically evolving on time scales comparable to those of the reactions they catalyze. Our goal is to improve catalytic activity and selectivity for dienes through fundamental understanding of the structure, dynamics, and catalytic properties of P-zeosils and through development of new, dynamically confined acids in zeosils and other microporous materials. Fundamentally, the description of weak acid catalysts requires new theoretical developments.

The synthesis of bio-renewable oleo-furan-sulfonate surfactants with unique properties will require development of a selective route for *direct* α-site acylation of a sugar-derived furan with carboxylic acids (derived from shale gas, sugars, fatty acids or triglycerides) to produce a 2-furyl alkyl-ketone. Fundamental understanding of catalyst design to promote *direct* acylation by fatty acids or triglycerides can provide significant molecular efficiency over the *indirect* route that uses fatty acid anhydrides, the production of which is a multi-step, waste-generating process. Use of bulky acylating agents in porous solid acids introduces new challenges and scientific questions regarding their distribution in porous materials and interaction with the active site(s), reaction mechanisms, and active site catalyst design. Pore design for the control of the distribution, orientation and diffusion of bulky amphiphiles within porous materials adds one more layer of complexity to be addressed. In turn, understanding these phenomena requires the development of computational, spectroscopic, and scattering methods.

Our endeavors into the design and synthesis of bio-lubricants with tunable molecular architecture and properties from versatile carbon feedstocks (biomass, natural oils and shale gas) will require precise control over branching with precursors of suitable size as well as control over oxygen content and ring saturation. To that end, we shall synthesize atomically-controlled catalysts that perform C-C coupling, hydrogenolysis of targeted furfural derivatives to produce alkylfuran substrates, and decyclization-hydrodeoxygenation for producing poly-alpha-olefin lubricants. Our goal is to develop new hydrogenolysis catalysts that exhibit simultaneously high activity and stability and to understand weak acidity in liquid phase chemistry. The multifunctional nature of these catalysts will require new advances in spectroscopic methods and use of cutting-edge imaging techniques combined with multiscale computational methods.

Catalysis Center for Energy Innovation (CCEI)	
University of Delaware	Dionisios G. Vlachos (Director), Eric Bloch, Stavros Caratzoulas, Yan Jin, Raul Lobo, Marat Orazov, Donald Watson, Bingjun Xu
Stony brook University / Brookhaven National Laboratory	Anibal Boscoboinik, Anatoly Frenkel
California Institute of Technology	Mark E. Davis
Carnegie Mellon University	Lynn Walker
Columbia University	Jingguang G. Chen
John Hopkins University	Michael Tsapatsis
Rutgers University	Marianthi Ierapetritou
University of California Santa Barbara	Song-I Han, Susannah Scott
University of Connecticut	Ioulia Valla
University of Maryland	Dongxia Liu
University of Massachusetts Amherst	Omar Abdelrahman, Wei Fan
University of Minnesota	Paul Dauenhauer
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Center for Molecular Magnetic Quantum Materials (M^2QM)
EFRC Director: Hai-Ping Cheng
Lead Institution: University of Florida
Class: 2018 – 2022

Mission Statement*: To provide the materials physics and chemistry understanding of molecular magnetic quantum materials essential for quantum and conventional computing beyond Moore's Law.*

Molecular magnetic materials (MMMs) are quantum materials, often with multiple exotic physical properties arising from the quantum spin states of the molecules and their coupling to the external environment and fields. The future of molecular magnetic systems (MMSs) as quantum material technology candidates depends critically on three fundamental scientific objectives: (i) mastering the chemistry of quantum magnets at the molecular scale in order to design and control all aspects of the energy states and their dissipation and dynamics; (ii) achieving the ability to design and make such materials as stable aggregates (crystals, adsorbates, nanostructures, etc.) while preserving or even enhancing their key physical properties; (iii) developing methods to interrogate and reversibly switch their quantum states (e.g. spin states). Making major advances towards these three critical capabilities is the primary goal for the Center for Molecular Magnetic Quantum Materials – M^2QM.

With an overarching goal of *turning molecular magnets into quantum materials useful for both quantum computing and quantum current conventional devices,* M^2QM aims to provide the materials physics and chemistry understanding needed to get beyond Moore's Law, *but with a failsafe strategic twist*. The Center's approach is equally targeted to both quantum devices (qubits) and quantum-current (e.g., spin) systems. For qubits, the MMS approach gives rigorous mono-dispersity (identical molecular magnets), enables genuine bottom-up assembly (including self-assembly) into complex aggregated systems, and enables electronic control and detection of quantum magnetic states by diverse means. For quantum currents, MMSs have inherently stronger coupling to the lattice than inorganic competitors (multiferroic oxides, transition-metal- dichalcogenides, etc). This allows the magnetic state to be sensed and manipulated more easily with low-energy electric or strain fields.

In the specific setting of this EFRC, molecular magnetic systems (MMSs) are molecular and nano-structured systems, including single molecule magnets, molecular-metal complexes, metal-organic quantum magnets, and counterpart multiferroics. MMSs exhibit immense spin configuration richness, spin crossover, or spin-state transition, all of which offer new ways to achieve multiferroic functionality. The distinction from materials phenomena rooted in the familiar quantum mechanical underpinnings of structure, bonding, and quasi-particle response illuminates the opportunity and challenge posed by quantum materials. The materials physics and chemistry must be right in two very distinct but intertwined ways: making a stable material from promising molecular constituents *and simultaneously enabling the emergence of the target phenomenon or phenomena (rather than causing its suppression).* Tasks in M^2QM are organized into three scientific thrusts as 1) Magneto-electric couplings in Quantum Materials, 2) Quantum Superposition and Entanglement of Molecular Spin States, 3) Spin-Charge Coupling on surfaces and interfaces, and a cross-cutting fundamental theory effort.

The "bottom-up" molecular/nanoscale approach (molecules → MMSs) to construction of spin logic devices long has been viewed as advantageous. Molecular structure can be designed to control low-energy electron/nuclear spin states. Indeed synthetic chemists now can design molecules with desired quantum states. As they are molecules, each is a precise replica of all others. Such replication obviously is critical for eventual scaling up to quantum devices with predictable performance. Templated substrates or scaffolds (such as polymers or nucleic acids) can attach and organize functionalized molecules, thereby providing routes towards development of quantum circuits. Other functionality can be built in, e.g., optically active transitions to provide fast sensing of the single-molecule electron spin state. Although information encoding and read-out can be done with the nuclear spin states of an individual molecule, that is inherently slow because of the low nuclear Larmor frequency in terrestrial magnetic fields. Thus electron spin qubits are vastly preferable. Other advantages of molecular electron-spin qubits versus some other qubit architectures are their relatively small size, monodispersity (uniform size), and the possibility of self-assembly into ordered crystalline ensembles. These advantages have allowed discovery of nebulous and fragile quantum properties in nanomagnetism that were difficult, if not impossible, to identify with traditional "top-down" magnetic nanoparticles. Those properties include quantum tunneling of the magnetization vector (QTM), followed by discoveries (involving members of this EFRC team) of exchange-biased QTM (EBQTM), quantum superposition states and entanglement (QSE), spin-spin cross relaxation (SSCR), and others. EBQTM, QSE, and SSCR were discovered in a hydrogen-bonded dimer of Mn_4 single-molecule magnets (SMMs) with spin $S = {}^{9}/_{2}$ ground states.

The monodispersity provided by bottom-up synthesis also enables very precise spectroscopic characterization of both the static and dynamic quantum properties of assemblies of magnetic molecules, something central to M^2QM. Beyond control over the physics of individual qubits, supramolecular design principles allow for connectivity between magnetic molecules and the incorporation of additional functionalities, such as optically active components that provide a means to switch and/or sense spin states at the individual molecule level. Other advantages include solution syntheses at or near room temperature, solubility in common solvents; formation of molecular crystals and structural characterization (to atomic precision) by single-crystal X-ray diffraction; and monolayer shells of organic ligands that can be varied conveniently to allow isotopic labeling (^{2}H, ^{19}F, etc). Additional merits include modulation of redox properties, crystal symmetry, and other properties.

M^2QM research will focus on the following topics: How intermolecular coupling changes structure and spin states of molecules via substitutions of ligands and linkers, and how such coupling is affected by pressure and fields; effect of coupling to substrate on the structure and magnetism of molecules and effect of molecules on the substrate such as the formation and variation of the Schottky barrier; spin state detection and manipulation; multiferroicity in molecular crystals manifested as coupling between spin state transition, strain, and charge polarization, in particular the magneto-electric effect; non-equilibrium dynamics in spin-phonon coupling; and effects of strong interaction and extreme fields. All studies will be carried out using combinations of experiment, theory, and computation.

Center for Molecular Magnetic Quantum Materials (M^2QM)	
University of Florida	Hai-Ping Cheng (Director), George Christou, Arthur Hebard, Richard Hennig, John Stanton, Neil Sullivan, Samuel Trickey, Xiaoguang Zhang
Florida State University	Stephen Hill (Deputy Director)
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Nanostructures for Electrical Energy Storage (NEES)
EFRC Director: Gary W. Rubloff
Lead Institution: University of Maryland
Class: 2009 – 2020

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 solid state nanostructures.*

In its first phase (2009-2014) NEES research was aimed at the design and electrochemistry of individual heterogeneous, multifunctional nanostructures as a pathway to high power energy storage together with stability during charge/discharge cycling. In its second phase (2014-2018) NEES-2 concentrated on architectural design, i.e. how to arrange precision nanostructures into dense mesoscale architectures for the same goals and to explore the new mesoscale science that emerges. In its current third phase, NEES-3 is focusing its full attention on all-solid-state electrochemical storage.

The opportunities in solid state storage (batteries and capacitors) are profound. Research on solid electrolytes and their electrode interfaces is miniscule compared to liquid-electrolyte systems, posing profound scientific challenges to understand the roles of chemical and electrostatic interactions at these interfaces. Advanced synthesis techniques from NEES play an enlarged role in solid state, enabling novel architectures that promise significant performance advantages and expanding potential applications beyond transportation and grid to biomedical, internet-of-things, and information storage for analog computation (Figures 1-3).

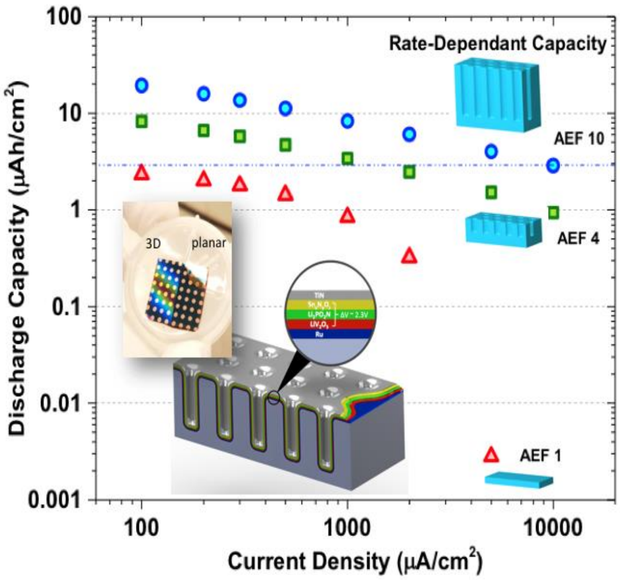

Figure 1. 3-D solid state battery enhances power and energy, shown here in comparison to planar (AEF 1) configurations.

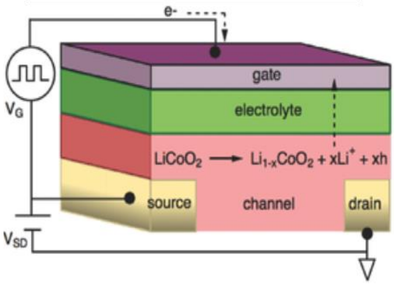

Figure 2. Solid state battery inspires nonvolatile redox memory based on ions stored in electrode

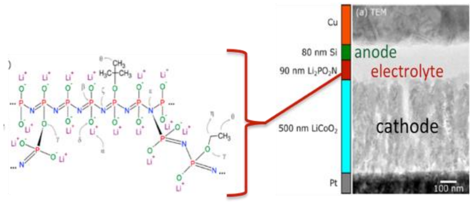

Figure 3. Highly controlled thin film synthesis (here ALD LiPON solid electrolyte) enables diverse 3-D architectures.

Many of the critical science questions are centered at interfaces, particularly that between solid electrolytes and electrodes, driving both theoretical efforts in computation (Figures 4-5) and experimental work in scanning probe microscopies and particularly for operando observations (Figures 6-7). Theoretical efforts are pursued at multiple length scales. Molecular-level DFT calculations reveal reaction energy barriers and likely kinetics. Continuum mechanics shows the changes in material properties induced by volume changes as ions are transported back and forth between electrode materials. Molecular dynamics provides insight into stochastic distributions that can dominate degradation and failure.

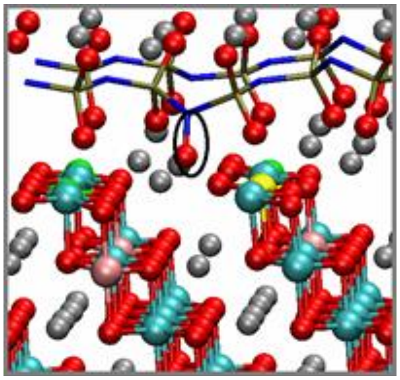

Figure 4. Model LiPON solid electrolyte is oxidized by $LiCoO_2$ cathode.

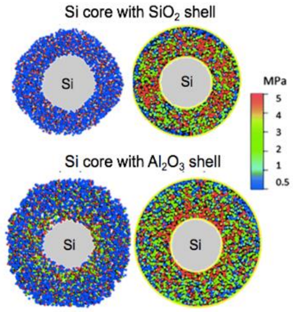

Figure 5. Stress evolution with lithiation/delithiation off core-shell nanostructures.

Experimental efforts are particularly aimed at understanding the complex energy landscape of critical regions in ionic solid state systems, specifically recognizing that the ions react according to chemical potentials but are sensitive to electrostatic (voltage) potentials during transport, while the reactions in which ions engage transform voltage contours across and near heterogeneous interfaces. Access to these and related issues is enabled by novel characterization approaches, including cross-sectional sample arrangements, patterned synthesis of multilayer structures, and localized electrochemical and mechanical configurations probed by operando SPM techniques.

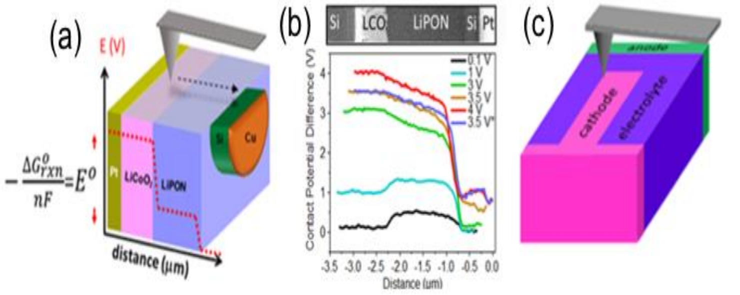

Figure 6. (a,b) KPFM voltage profile across solid interfaces; (c) KPFM operando imaging in 3D interdigitated solid state battery.

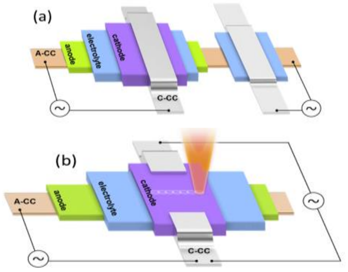

Figure 7. Patterned thin film testbeds to characterize solid state battery electrochemistry (a) and to identify specific energy barriers at interfaces as a function of charge state.

The potential scientific impact in fundamental understanding of all-solid-state electrochemical storage is profound, attracting a growing community of researchers, with important and diverse applications in sight. NEES looks forward to expanding its current leadership in this research domain.

Nanostructures for Electrical Energy Storage (NEES)	
University of Maryland	Gary Rubloff (Director), Sang Bok Lee (Deputy Director), Janice Reutt-Robey
Sandia National Laboratories	A Alec Talin, Kevin Leung
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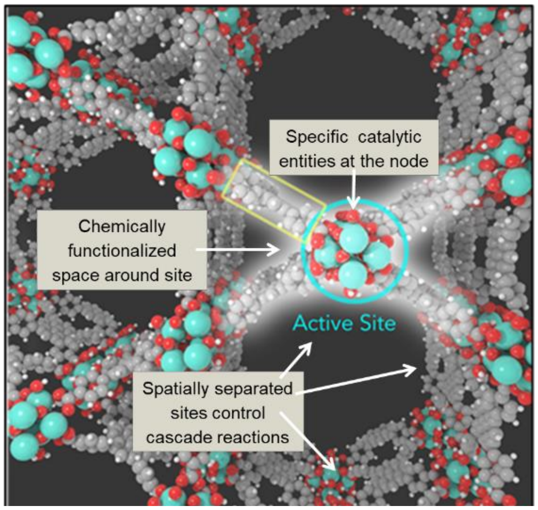
Inorganometallic Catalyst Design Center (ICDC)
EFRC Director: Laura Gagliardi
Lead Institution: University of Minnesota
Class: 2014 – 2022

Mission Statement*: To discover new classes of energy-science relevant catalytic materials, especially through the exploitation of computational modeling to identify underlying structure-function relationships that are critical to advancing further, predictive catalyst discovery.*

ICDC pursues hypothesis-driven experimental, computational, and theoretical research in heterogeneous catalysis. Our **experimental vision** is a) to devise and apply synthetic methods to yield stable, uniformly organized arrays of catalytic sites with single-atom or close-to-single-atom precision with respect to both composition and placement, and b) to use these catalysts to answer fundamental questions in catalysis science. Paramount to our approach is the synthetic accessibility of chemically and thermally stable metal-organic frameworks (MOFs), featuring controlled porosities, ultra-high surface areas, and well-defined catalytic sites, or anchoring sites for catalysts, along with tailored environments around such sites.

Our **vision for theory and computation** is to develop novel quantum chemical, molecular mechanical, and combined quantum mechanical and molecular mechanical approaches to explore the structures, stabilities, activities, and selectivities of realized and as-yet-unrealized catalytic materials, where the latter can be proposed as targets for synthesis by the experimental teams in the Center. Using transferrable advances in theory developed by ICDC, we augment detailed characterizations of elementary reaction steps with microkinetic modeling to identify key descriptors that can be used to rapidly screen potential catalysts and tune catalyst behavior.

Central to our efforts is the use of frameworks based on hexa-zirconium(IV) nodes and oxy-anion terminated linkers. $Zr^{4+}O^{2-}$ bonds are among the strongest known ionic bonds. Oxyanion-terminated linker-node bonds are not so strong but are among the strongest known for MOFs. Known members of this MOF family exhibit especially desirable properties for catalysis science and technology.



Schematic representation of a catalytically active site in hexa-zirconium(IV)-node based MOF and the elements critical to constituting this site.

To address the most challenging and compelling questions at the forefront of catalysis science, **the mission of ICDC** will be to develop and promulgate:

- Concerted experimental/computational approaches to (i) make and interpret structural and spectroscopic observations, (ii) measure, interpret, and predict catalytic properties, and (iii) guide new MOF syntheses.
- Experimental methods for rapid, robust synthesis of previously unexplored, well-defined, catalytic structures in MOFs—including those with compositions unattainable on the macroscopic scale.
- Theoretical models able to guide experiment for discovery and rationalization of structure–function relationships in catalysts incorporating inorganometallic structures ranging from single metal atoms to clusters smaller than bulk-like nanoparticles.

Having demonstrated the reproducible synthesis of many catalytically active sites embedded in the three-dimensional environments of MOFs, we are now positioned to design novel catalysts that master the complexity of catalyzed transformations, guided by computational insights into elementary steps and enabled by advanced synthesis. To achieve this goal we will exploit: (i) confinement of the locations of active sites; (ii) nuclearity of active centers; (iii) tailored chemical environments surrounding centers; (iv) controlled under-coordination (defects) to create new access to key MOF elements; and (v) steric separation of sites with different functions to enable multiple reaction steps.

Focusing on the catalytic chemistry of shale-gas substrates, we will address the selective conversion of light alkanes, including methane, as well as the synthesis of fuel and chemical components from light alkane conversion. The target catalytic transformations will be those that can best answer important, fundamental questions in energy-relevant catalysis science and in energy-relevant materials synthesis. At the highest level, these catalytic transformations will address the fundamental challenges of controllably making and breaking C-H, C-C, C-O, C-N, and C-S bonds, as well as bonds between these various atoms and metal or metal-oxide catalysts.

We envision five groups of deliverables from our program:

- Advanced computational methods and strategies to aid interpretation of mechanistic information, as well as structural and spectroscopic observations to guide synthesis.
- Advanced methods to synthesize electronically and structurally defined catalytically active sites in heterogeneous catalysts with unprecedented precision.
- Methods for precisely siting active sites in mesoscale, uniform, and hierarchically structured environments that both isolate sites and provide ready access to them.
- Advanced characterization methods of (operando) catalyst function and structure.
- Stringent approaches for the discovery and theoretical rationalization of structure/function relationships in atomically precise catalysts in cluster size ranges of less than 100 atoms.

Research Approach. It has been a central tenet of ICDC that theory would not only help to post facto interpret spectroscopic and chemical observations, but that it would also guide experimental priorities, as the nearly infinite number of possibilities imposes need for a hypothesis-based, step-by-step approach to catalyst development. Our ongoing hypotheses are informed by observations from ICDC and current, preliminary experiments. Theory and physicochemical evaluations of catalysts and their reactions have synergistically provided the tools to understand their chemical and structural properties, and we see substantial promise for making rational future predictions.

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Alliance for Molecular PhotoElectrode Design for Solar Fuels (AMPED)
EFRC Director: Gerald J. Meyer
Lead Institution: University of North Carolina at Chapel Hill
Class: 2009 – 2020

Mission Statement*: To develop the fundamental molecular basis for solar-driven water oxidation and carbon dioxide reduction catalysis.*

Sunlight is the one renewable natural resource that could by itself meet all the energy demands of our growing world economy. For solar energy to reach its full potential it must be coupled to a storage method, which can be achieved by conversion of sunlight into chemical energy stored in bonds of molecules called solar fuels. Inspired by natural photosynthesis, the mission of the **A**lliance for **M**olecular **P**hoto**E**lectrode **D**esign for Solar Fuels (AMPED) is to *develop the fundamental molecular basis for solar-driven water oxidation and carbon dioxide reduction catalysis*. This mission will be achieved by understanding how molecular catalysts and chromophores can be integrated covalently and non-covalently with oxide materials to achieve structurally well-defined photoelectrodes that efficiently couple light absorption with multi-electron fuel-forming reactions. Two integrated research goals that support the AMPED EFRC mission and specifically address pressing knowledge gaps in energy science are proposed:

1. **Interface Highly Conductive Oxides with Molecular Light-Harvesting Dyes and Photocatalysts.**

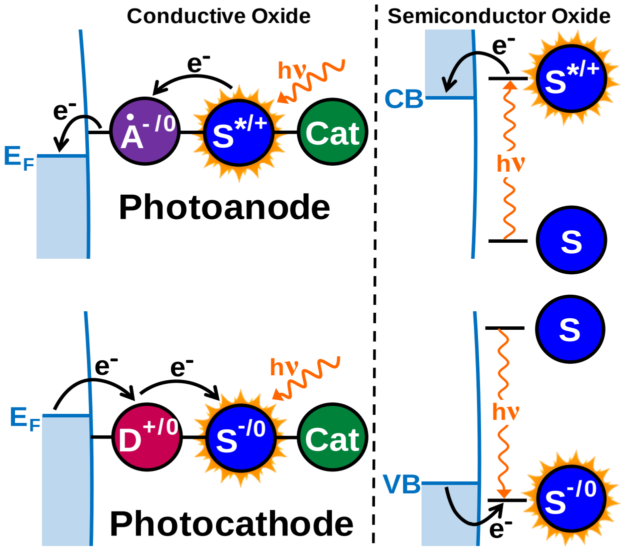

Figure 1. Dye-sensitized conductive (left) and semiconducting (right) oxide photoelectrodes.

A new approach to highly efficient light-driven catalysis using simple *conductive* oxides offers a promising alternative to the prevailing use of insulating or semiconducting oxides. Fundamental studies will elucidate dye-sensitized electron transfer reaction mechanisms and guide the development of new photoelectrode designs. Highly doped transparent conductive oxide (TCO) nanocrystalline mesoporous thin films will be utilized for dye-sensitized solar fuel generation. Recent AMPED EFRC research has demonstrated that, with careful design of the molecule-TCO interface, productive electron transfer reactivity can be realized that enables photocatalysis. In such studies the dye excited state undergoes rapid electron transfer with a molecular donor or acceptor — thereby precluding deactivation by the conductor (Figure 1). With an applied potential, the TCO has been shown to fulfill the role of an *n*- or *p*-type semiconductor material by collection of reducing or oxidizing equivalents. This breakthrough discovery changes the way we think about materials, with an emphasis on the position of the Fermi level, E_F, of the oxide material under solar irradiance conditions (whereas the band edge position is the key parameter for semiconductors). Detailed experiments will enable molecular control of interfacial dynamics and catalysis at conductive oxides by determining reorganization energies and reduction potentials associated with electron transfer. Preliminary results have demonstrated both remarkably long-lived charge separation and multi-electron/multi-proton catalysis.

2. **Direct Multi-Electron Flow at Photocatalytic Semiconductor Electrodes.**
Synthetic photoelectrochemistry for solar fuel production requires that light absorption be efficiently coupled with chemical bond formation, achieved through the directed flow of electrons and protons *away* from water oxidation catalysts and *towards* CO_2 reduction catalysts, Figure 2. Electron flow has been optimized in prior UNC EFRC research through the spatial arrangement of redox active centers or band edge positions that provide a free energy gradient. Methods for directional transfer of both electrons *and* protons do not exist, yet are critically important at oxide interfaces. Interfaces will be designed to direct proton and electron flow both *between* interfaces and also *laterally* within a component layer. Unlike the TCOs in Goal 1, catalysis generally occurs at potentials within the forbidden energy gap, conditions where the oxide acts as an insulator and blocks electronic communication with the external circuit. The semiconducting oxides will be exploited to characterize the redox behavior of site-isolated catalysts so as to control intermolecular reactions. The interfacial architectures will be quantified in aqueous solution using time-resolved spectroscopic and electrochemical techniques. By developing synthetic methods that provide spatial control of molecule-material interfaces, and tuning electronic and protonic coupling at interfaces, we aim to identify and understand the basic principles that direct flow between dyes, catalysts, and semiconductor oxides in the dark and under solar illumination.

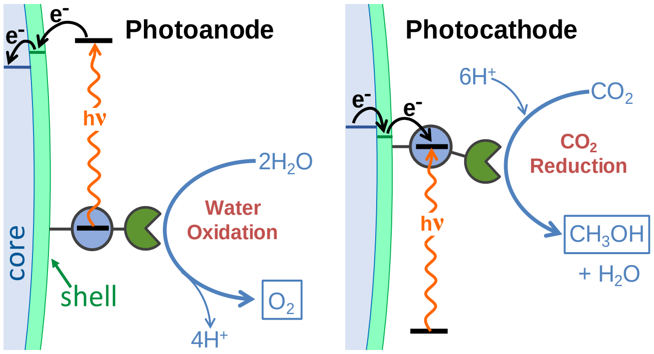

Figure 2. Desired flow of electrons and protons at dye-sensitized semiconductor interfaces.

An approach rooted in comprehensive mechanistic analysis, at the molecular level and at molecule-materials interfaces, will establish new paradigms in light-driven fuel synthesis based on exquisite control over photoelectrode structure and dynamics. AMPED EFRC research will leverage expertise in molecular and materials synthesis and characterization, catalysis, time-resolved spectroscopies, kinetics and modeling, and photo-electroanalytical methods. The proposed multi-disciplinary team-based approach has proven to be highly effective in bringing together a broad spectrum of ideas and capabilities necessary for transformational advances in solar fuels research that could not be realized by individual research groups. The impact of AMPED EFRC research will be a deeper mechanistic understanding of sunlight-driven catalytic transformations at molecule-material interfaces, which will in turn fuel the discovery of new breakthroughs in solar energy conversion chemistry.

Alliance for Molecular PhotoElectrode Design for Solar Fuels (AMPED)	
University of North Carolina at Chapel Hill	Gerald J. Meyer (Director), Alexander J. M. Miller (Deputy Director), Joanna M. Atkin, Maurice S. Brookhart, James F. Cahoon, Jillian L. Dempsey, Yosuke Kanai, Thomas J. Meyer, John M. Papanikolas, Cynthia K. Schauer, Joseph L. Templeton, M. Kyle Brennaman
University of Texas at San Antonio	Kirk S. Schanze
Georgia Institute of Technology	John R. Reynolds

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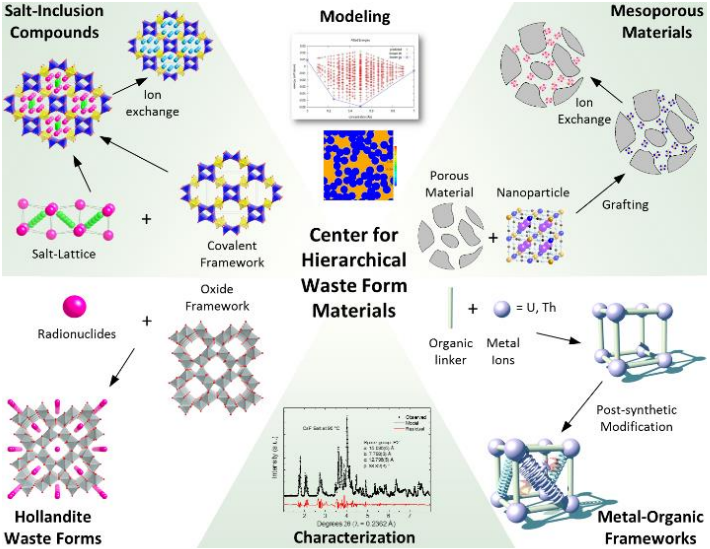
Center for Hierarchical Waste Form Materials (CHWM)
EFRC Director: Hans-Conrad zur Loye
Lead Institution: University of South Carolina
Class: 2016 – 2020

Mission Statement*: To combine experiment and modeling to develop the chemistry and structure motifs needed to create hierarchical materials that effectively immobilize nuclear waste in persistent architectures.*



The Center for Hierarchical Waste Form Materials (CHWM) EFRC is organized to develop the basic science and foundational knowledge from which new waste forms can emerge. The center combines synthesis, characterization and modeling to develop and validate chemistry and structure motifs for materials that effectively immobilize nuclear waste materials into the indefinite future. Novel hierarchical structures are being developed via a bottom–up synthetic approach, where new chemical insights provide the required critical understanding of fundamental mechanisms of complexation and speciation. The outcome will be the advancement of fundamental knowledge related to hierarchical materials systems and their potential future application in waste forms.

A simple definition of a hierarchical material is that of a structural motif contained within a larger structure or framework. Conceptually, the hierarchical structures consist of porous frameworks, either repeating (crystallographically ordered) or non-repeating (disordered), whose cavities can be occupied by crystalline or non-crystalline fillers. Examples of these structures include porous crystalline salt inclusion materials (SIMs), metal-organic frameworks (MOFs), porous silica (including Prussian-blue analogues (PBA) and silver salt functionalized versions), and surface functionalized nanoparticles assembled into hierarchical constructs. Examples of fillers include crystalline salt phases, simple molecular species, silver salt nanoparticles, and Prussian-blue nanoparticles, all either freely located inside the pores or tethered/bonded to the pore walls.

The effort is supported by extensive modeling across multiple length and time scales. Ab initio modeling of alloy nanoparticle stability is utilizing derived Charge Optimized Many Body (COMB) potentials, and simulating atomic-scale processes using tools such as molecular dynamics. Thermochemical models are being developed for complex phases, such as the SIMs, that allow prediction of stability, vapor pressure, melting point, including prediction of the pore filling species, using density functional theory and by extending approaches such as topological and volume-based correlations. At the mesoscale, phase field methods are

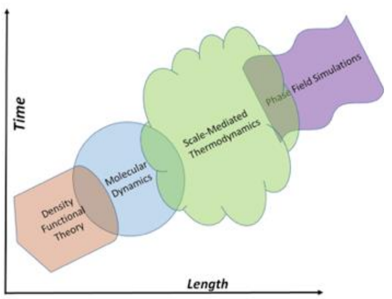


Illustration of bridging scales in modeling waste forms.

being generated for the simulation of porous solids using datasets of surface energies, thermodynamic models, and configurational thermodynamics.

Sample characterization takes advantage of in-laboratory diffraction equipment at Alfred University, state of the art calorimetry measurement systems at Clemson University, thermal analyzers at USC, and beam line facilities, including the SNS at ORNL, and the APS at ANL, for in-situ crystallization, ion-exchange, and general structural characterization. This integrated research is creating the science that will eventually lead to functional, complex material structures for efficient and effective waste sequestration. In addition, access to Savannah River National Laboratory's (SRNL's) 773-A category 2 nuclear facility will enable us to create radioactive versions of hierarchical structures, in addition to the surrogate versions, so as to better understand the unique properties of forms containing elements such as technetium.

The CHWM focuses on a number of hierarchical structures, including SIMs, MOFs, and multi-scale porous silicate structures, developing advances in synthesis, measurement of ion exchange properties, modeling of ion exchange pathways, predicting and measuring thermochemical stabilities, and applying advanced characterization tools, often in novel ways, such as synchrotron and neutron sources, to create and understand hierarchical materials that can lead to the effective immobilizing nuclear waste in persistent architectures. The cross cutting tasks effected by the use of working groups encourage information exchange between center personnel, fostering synergy between the groups, and assure that members are working toward the Center's overarching goals. Specifically, the methodology for synthesizing new uranium containing SIMs has been achieved and has allowed the Center to prepare a large number of novel frameworks, including those based on new building blocks, such as the first $[Al_2O_7(PO_3)_6]^{14-}$ secondary building unit (SBU) in uranium chemistry. The work on multi-scale porous silicate structures has resulted in the preparation of silica monoliths with multi-scale porosity, where optimization of the synthetic process has resulted in the formation of centimeter-scale silica samples with an open macro and mesoporosity. The introduction of PBA nanoparticles inside the structure was achieved by in-structure synthesis of PBAs, which enables the entire sorption capacity of the PBA particles to be used. Another class of hierarchical structures under investigation are MOFs containing U and Th that have been prepared as part of the Center work. New synthetic routes for U and Th incorporation were developed, including metal node extension, post-synthesis modifications and transmetallation. The synthesis of new linkers and capping ligands has been achieved with the goal to trap species inside the MOFs. The hollandite structure, $A_xM_8O_{16}$, is being investigated as a potential candidate for the immobilization of alkali metal radionuclides such as Rb and Cs. The chemical flexibility of the hollandite structure leads to varying degrees of structural disorder that modeling is addressing

Center for Hierarchical Waste Form Materials (CHWM)	
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Savannah River National Laboratory	Jake Amoroso, David DiPrete, Charles Crawford
Pacific Northwest Laboratory	Charles Henager, Shenyang Hu, Yulan Li
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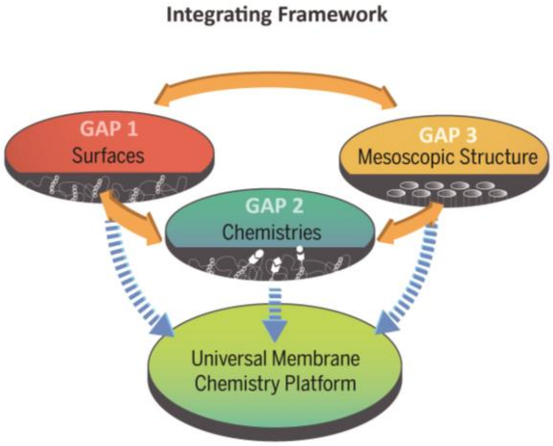
Center for Materials for Water and Energy Systems (M-WET)
EFRC Director: Benny D. Freeman
Lead Institution: The University of Texas at Austin
Class: 2018 – 2022

Mission Statement*: To discover and understand fundamental science to design new membrane materials, develop tools and knowledge to predict new materials' interactions with targeted solutes from recalcitrant water sources, provide fit for purpose water, and recover valuable solutes with less energy.*

Contaminated water from energy-related activities is an enormous burden (wastewater management) and an exciting untapped opportunity (resource recovery). Synthetic membranes are widely used for purifying relatively clean water due, in part, to low energy requirements of membranes relative to alternatives (e.g., thermally based separations). However, today's membranes were not designed to treat highly impaired water, such as produced water, due to extensive fouling and poor separation properties. Existing membranes: (1) are poor at discriminating between ions of the same valence (e.g., Na^+ v. Li^+), (2) have low selectivity for many neutral contaminants (e.g., boron, arsenic), (3) are always subject to fouling, (4) exhibit a pernicious tradeoff between permeability and selectivity, and (5) are produced in poorly understood, highly non-equilibrium processes that limit deliberate control of their properties. Fundamental knowledge gaps contributing to these shortcomings and frustrating membrane design include: (1) structure/dynamics of hydration water and solutes (e.g., ions, dissolved organics, etc.) near membrane/fluid interfaces, in membrane separation layers, and in pores, (2) thermodynamic and kinetic properties of solutes in aqueous mixtures, near membrane/fluid interfaces and in membranes that depend, in part, on solute and surface hydration properties, and (3) rational design of selectivity-enhancing interactions between water, solutes and membranes.

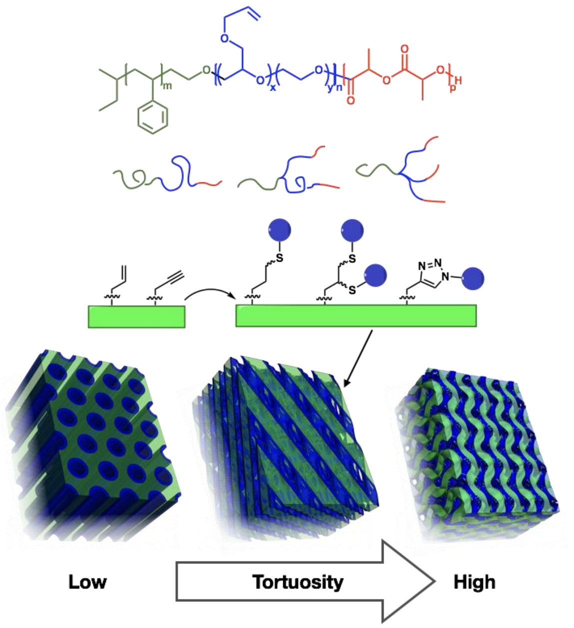
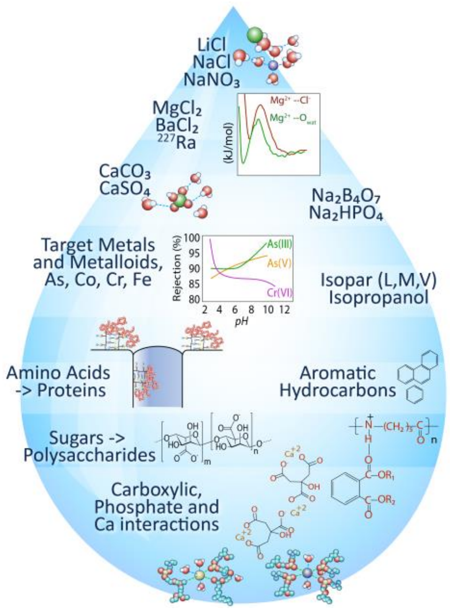
The Center for Materials for Water and Energy Systems (M-WET) will bridge the chemistry, materials and process separation communities to: (1) design new interfaces to achieve optimal affinity and reactivity for water/energy systems (e.g., ion-specific separation); (2) precisely control mesoscopic materials architecture to achieve exquisite control of pore size and pore size distribution in membranes; (3) develop novel materials imaging and spectroscopic tools that operate in-situ/in operando in complex, aqueous fluid environments to probe water, solute and material interactions; and (4) model interfaces, separation membranes, fluid mixtures, and mesoporous architecture to radically transform water and energy demands, resiliency, and efficiency of membrane/materials systems.

M-WET comprises three Gap Attack Platforms (GAPs) focused on: (1) molecular design of surfaces to control and tune water properties at interfaces; (2) designing specific interactions to improve membrane separation properties; and (3) mesoscale structures to tailor fluid flow through porous and isoporous membranes. A cross-cutting Integrating Framework leverages these materials design insights to provide directions for breakthrough improvements in real separation processes. Achieving these goals requires deep, sustained, and interdisciplinary efforts in synthesis, characterization, and modeling.



M-WET scientific framework, illustrating the Gap Attack Platforms (GAPs) and Integrating Framework.

A single, modular model materials platform, the Universal Membrane Chemistry Platform (UMCP), is used and functionalized in GAPs 1–3, so breakthroughs are seamlessly transferred among GAPs and across length scales. The GAPs also share a Model Fluid Platform (MFP) to provide continuity, coherence, and relevance among research projects. The MFP comprises a hierarchy of increasingly complex fluids for use across all GAPs, beginning with water, water + simple salts, water + organics (e.g., dissolved organics or emulsified oil or both), water + salt + organics, and ultimately, model produced water containing organic and inorganic components.

Universal Membrane Chemistry Platform (UMCP)	**Model Fluid Platform (MFP)**
	

M-WET's Universal Membrane Chemistry Platform and Model Fluid Platform.

Materials for Water and Energy Systems (M-WET)	
The University of Texas at Austin	Benny Freeman (Director), Lynn Katz (Associate Director), Nate Lynd, Tom Truskett, Venkat Ganesan, Des Lawler, Mukul Sharma, Michael Webber
University of California, Santa Barbara	Rachel Segalman (Associate Director), Mahdi Abu-Omar, Chris Bates, Mike Doherty, Glenn Fredrickson, Songi Han, Craig Hawker, M. Scott Shell, Todd Squires
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Multi-scale Fluid-Solid Interactions in Architected and Natural Materials (MUSE)
EFRC Director: Darryl Butt
Lead Institution: University of Utah
Class: 2018 – 2022

Mission Statement*: To synthesize geomaterials with repeatable hierarchical heterogeneity and develop an understanding of transport and interfacial properties of fluids confined within these materials.*

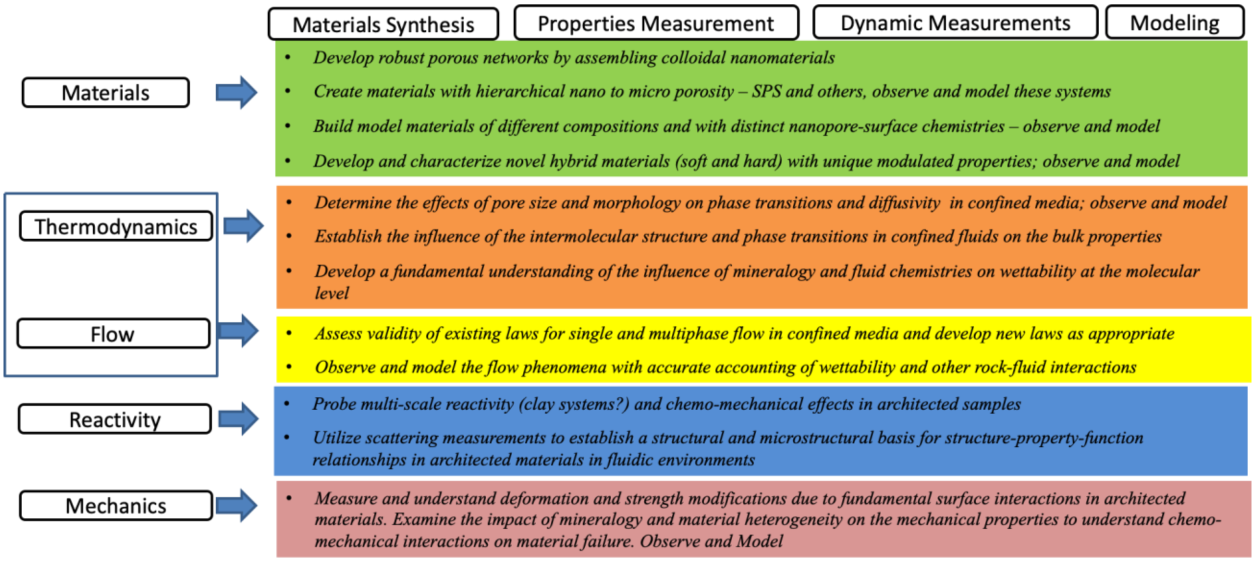
Successful operations of energy recovery and storage, sensor technologies, membranes for air and water purification, and catalytic processes require a fundamental understanding of the interactions of fluids at solid interfaces. There is considerable evidence that the known laws of adsorption, reaction, phase transitions, flow and mechanical strength are affected by the presence of fluids confined in porous geomaterials with nanometer-sized pores.

To accomplish transformational changes in energy science, MUSE will 1) develop a fundamental understanding of confinement and surface interactions in geomaterials on fluid phase behavior, reactivity, and multiphase flow properties; 2) examine the impact of mineralogy and material heterogeneity on chemo-mechanical properties; 3) determine *in-operando* cross-scale structural and microstructural material properties with fluids in confinement; and 4) develop validated multiscale and multi-physics models of mechanics, phase behavior, and flow that capture the observed chemo-morphological coupling.

MUSE is organized into four functional *Research Thrust Areas*: (1) Materials Architecture and Characterization, (2) Property Measurements, (3) Dynamic Characterization, and (4) Multiscale Multiphysics Modeling, and five, cross-cutting scientific areas as highlighted in Figure 1. Novel geo-architected materials developed in Thrust 1 are used as substrates for determining properties of multi-phase fluids in heterogeneous confined architectures in Thrust 2, and for dynamic *in-operando* determination of material and fluid properties in Thrust 3. These measurements are informing the development of *experimentally-validated*, *atomistically-informed* modeling tools and frameworks in Thrust 4.

The functional thrusts interact with members across the center to address scientific themes and technical challenges as summarized in Figure 1. Hierarchical nanostructured geomaterials with increasing levels of complexity are being created and characterized. Experiments with similarly–ordered, naturally occurring materials such as shales will also be performed for comparison. Phase behavior, flow and mechanical properties measurements will demonstrate chemo-mechanical interactions. These new phase transition measurements in architected geomaterials at realistic conditions provide confirmation of modeling findings. In-operando observation of such phenomena will generate enhanced understanding of how confinement and surface interactions are affecting these fluid properties.

The Dynamic Characterization focus area will develop *in situ (in-operando)* observation methods at system conditions. X-ray and neutron radiation provided by Basic Energy Sciences (BES) facilities allows the research team to probe the structure and dynamics of matter spanning from the molecular to the macro length scales. Detailed nonreactive and reactive molecular dynamics (MD) modeling, enhanced by first principles quantum mechanics calculations, will be applied to improve fundamental understandings of, 1) phase behaviors of multicomponent fluids confined in nanometer size pores, 2) heterogeneous adsorption of fluid molecules on the heterogeneous surfaces of organic and inorganic materials, and 3) diffusion of fluid molecules within nanopores, and 4) reaction-induced mechanical deformations of nanostructures.



- *Develop robust porous networks by assembling colloidal nanomaterials*
- *Create materials with hierarchical nano to micro porosity – SPS and others, observe and model these systems*
- *Build model materials of different compositions and with distinct nanopore-surface chemistries – observe and model*
- *Develop and characterize novel hybrid materials (soft and hard) with unique modulated properties; observe and model*
- *Determine the effects of pore size and morphology on phase transitions and diffusivity in confined media; observe and model*
- *Establish the influence of the intermolecular structure and phase transitions in confined fluids on the bulk properties*
- *Develop a fundamental understanding of the influence of mineralogy and fluid chemistries on wettability at the molecular level*
- *Assess validity of existing laws for single and multiphase flow in confined media and develop new laws as appropriate*
- *Observe and model the flow phenomena with accurate accounting of wettability and other rock-fluid interactions*
- *Probe multi-scale reactivity (clay systems?) and chemo-mechanical effects in architected samples*
- *Utilize scattering measurements to establish a structural and microstructural basis for structure-property-function relationships in architected materials in fluidic environments*
- *Measure and understand deformation and strength modifications due to fundamental surface interactions in architected materials. Examine the impact of mineralogy and material heterogeneity on the mechanical properties to understand chemo-mechanical interactions on material failure. Observe and Model*

Figure 1: The four functional and scientific thrust areas of MUSE highlighting the major cross-cutting technical challenges that are being addressed.

The Center is generating new data and creating new models on phase transitions, flow characteristics, and multiphase properties in confined geomaterials. MUSE brings together a multi-disciplinary team to establish a multi-scale scientific basis for advancing energy technologies that are of critical importance to the current and future world energy security and environmental sustainability.

Multi-scale Fluid-Solid Interactions in Architected and Natural Materials (MUSE)	
Idaho National Laboratory	Yidong Xia, Joshua Kane
The Pennsylvania State University	Adri van Duin (Thrust 4 Lead)
University of California, Davis	Subhash Risbud
University of Utah	Darryl Butt (Director), Michael Bartle (Deputy Director), Milind Deo (Deputy Director), Michael Hoepfner Jules Magda, John McLennan Brian McPherson, Jan Miller (Thrust 2 Lead), Swomitra Mohanty, Pania Newell, Bryony Richards, James Sutherland, Ilya Zharov (Thrust 1 Lead)
Cornell University	Greeshma Gadikota (Thrust 3 Lead)
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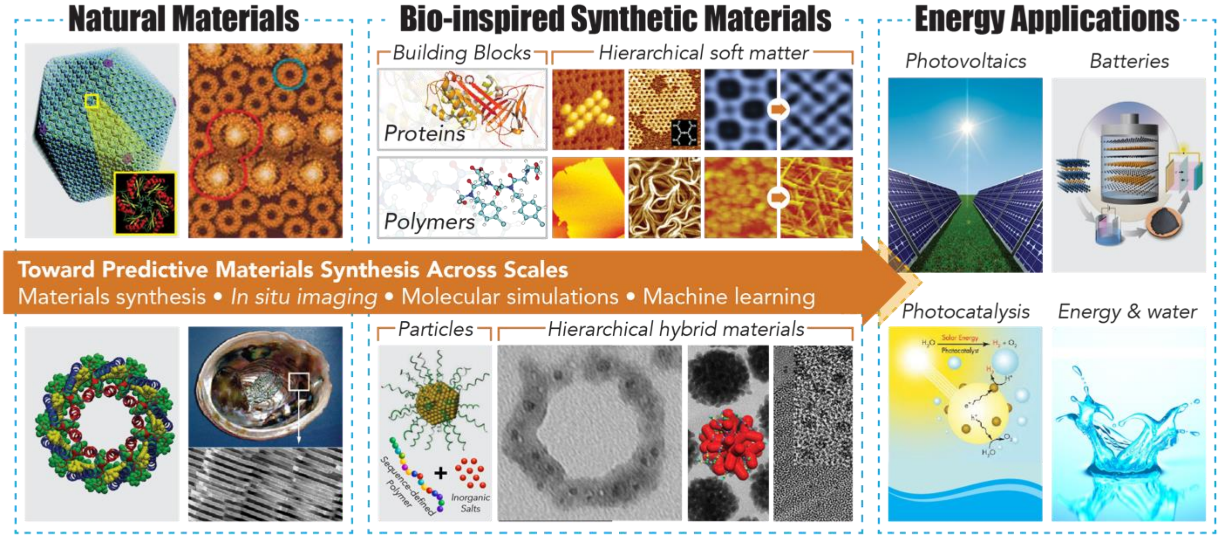
The Center for the Science of Synthesis Across Scales (CSSAS)

EFRC Director: François Baneyx
Lead Institution: University of Washington
Class: 2018 – 2022

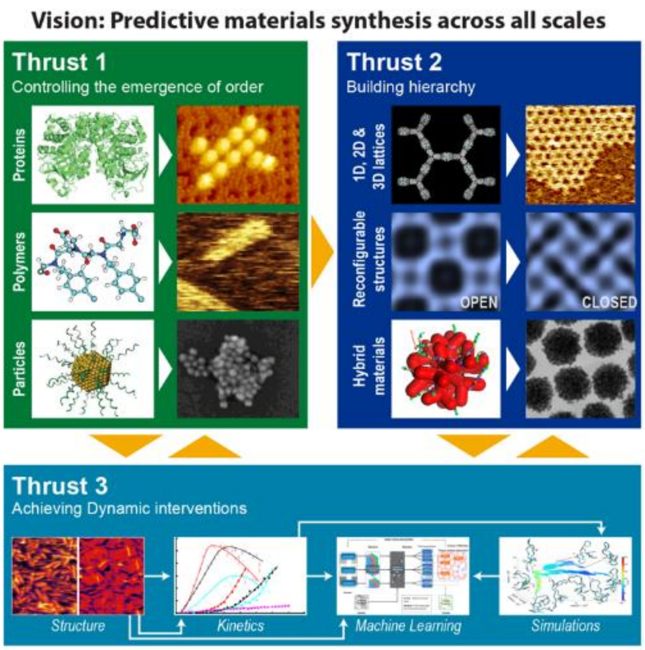
Mission Statement*: To harness the complex functionality of hierarchical materials by mastering the design of high-information-content macromolecular building blocks that predictively self-assemble into responsive, reconfigurable, self-healing materials, and direct the formation and organization of inorganic components.*



Hierarchical materials offer game-changing opportunities for energy technologies because they enable complex interconnected functions ranging from control of charge and mass transport, to dissipative response to external stimuli and the precise localization of sequential and parallel reactions. Nowhere is this more evident than in nature where hierarchical materials perform the stunning range of functions that has made life possible. While there have been many success stories in the quest to synthesize biomimetic and bioinspired materials with outstanding structure and function, efforts have not scratched the surface of what is possible because they have been driven by intuition and serendipity rather than by a deep predictive understanding of the fundamental rules underpinning hierarchical materials synthesis. We have created CSSAS to realize a shared vision: mastering the design of high-information-content macromolecular building blocks that predictively self-assemble into hierarchical materials. Currently, four major knowledge gaps stand between us and that vision:

1. We have little knowledge of how sequence and chemistry translate into molecular interactions and assembly dynamics from which order emerges.
2. We cannot yet connect atomistic descriptions of intermolecular interactions with coarse grained models of building blocks to bridge the time and length scales required for predicting assembly.
3. We do not know how the interplay of intermolecular interactions, solvent and electrolytes, disparate building blocks, and dynamic processing conditions, controls energy landscapes across which hierarchy develops.
4. We do not know how to predict metastable states on the pathway to the final ordered state, or how to encode a balance of forces that will, by design, create multi-well potentials for out-of-equilibrium switching in response to external stimuli.

With a highly synergistic team of internationally recognized thought-leaders from the University of Washington (lead institution), Pacific Northwest National Laboratory, the University of Chicago, Oak Ridge National Laboratory, and the University of California San Diego, CSSAS will fill these knowledge gaps by tackling three scientific goals:



1. To predict how the chemical and/or sequence information of inorganic, polymer and protein building blocks translates into the emergence of order and the outcomes of assembly.

2. To master the free energy landscapes of disparate building blocks in complex environments and at surfaces to control their assembly into (functional and reconfigurable) hierarchical units, as well as the morphogenesis and organization of inorganic components.

3. To integrate the tools of data science with in situ characterization, and simulations to achieve adaptive control of synthetic outcomes and access metastable states of matter.

Our hypothesis-driven research plan tackles the first goal by creating a set of systematically variable building blocks that span the scale of complexity – from large proteins to atomically-precise inorganic clusters – and by combining *in situ* observations with a hierarchy of simulation techniques that describe interactions and predict how order emerges. Our plan accomplishes the second goal by exploring the frontier of integration and hierarchical assembly of building blocks, while extending observations and simulations to length and timescales where hierarchy comes into full bloom. Finally, our plan addresses the third goal by exploiting the richness of *in situ* data and the predictive capacity of molecularly-informed coarse graining to harness the power of data-driven machine learning, where the full potential of real-time datasets is enlisted through data analytics. In doing so CSAAS will bridge the key knowledge gaps in the field biomolecular materials and create a lasting scientific foundation that advances BES's priority research directions, grand challenges and transformative opportunities.

The Center for the Science of Synthesis Across Scales (CSSAS)	
University of Washington	François Baneyx (Director), David Baker, Brandi Cossairt, David Ginger (Thrust 3 Lead), Sam Jenekhe, Jim Pfaendtner (Thrust 2 Lead)
Pacific Northwest National Laboratory	Jim De Yoreo (Deputy Director), Chun-Long Chen, Chris Mundy (Thrust 1 Lead)
University of Chicago	Andrew Ferguson
University of California San Diego	Akif Tezcan
Oak Ridge National Laboratory	Sergei Kalinin

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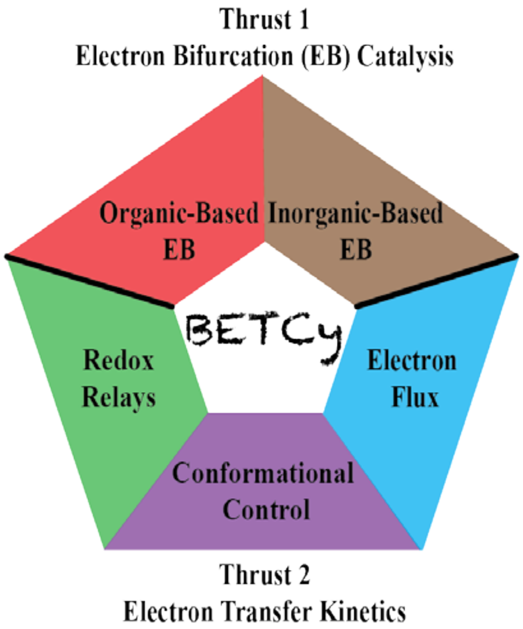
Biological Electron Transfer and Catalysis Center (BETCy)
EFRC Director: John Peters
Lead Institution: Washington State University
Class: 2014 – 2020

Mission Statement*: To understand the means by which biology controls the kinetics and thermodynamics of electron bifurcation at both organic and inorganic centers through electron transfer relays, allosteric coupling, and cooperative conformational dynamics.*

The focus of the Biological Electron Transfer and Catalysis (*BETCy*) EFRC research is elucidating mechanisms of conversion of electrochemical potential into chemical bond energy and is organized into two integrated Thrusts: Electron Bifurcation Catalysis and Electron Transfer Kinetics.



We are developing a collective knowledge of metalloenzymes as models for redox reactions by applying physical science and computational tools to characterize biochemical reactions catalyzed by multi-subunit enzymes harboring arrays of iron-sulfur clusters and flavin cofactors. Understanding these mechanisms is central to overcoming the thermodynamic barriers that currently limit production of reduced products and fuels.

Biological systems have elegant strategies for converting electrochemical potential energy into chemical bond energy (e.g., C-H, H-H, and N-H) stored in reduced compounds that can serve as advanced biofuels. One significant limitation for the production of highly reduced compounds is that their production in natural and industrial processes relies on low oxidation-reduction potential chemicals as feedstocks. However, unique biochemical strategies exist to generate pools of reducing equivalents that can serve as a source of electrons for chemical bond formation from low oxidation-reduction potential feedstocks.

We are focusing on a newly discovered biochemical mechanism termed “electron bifurcation”, which upgrades electrochemical potential by effectively coupling endergonic and exergonic reactions in an overall thermodynamically favorable process. The underlying mechanistic details governing electron bifurcation are, however, still poorly understood and a more in-depth understanding of this phenomenon could lead to “game changing” and transformational advances in strategies to direct electron flow. These studies provide a blueprint for bio-inspired, multi-electron catalytic processes that can ultimately utilize electrons of varying reduction potentials to drive chemical reactions. We are also working to elucidate how biology uses a combination of chemical bond energy and electrochemical potential to accomplish very difficult, low potential reduction reactions (e.g., CO_2 and N_2 reduction) using electron donors of modest reduction potentials.

The goal of the *BETCy* EFRC is to provide a fundamental understanding of mechanisms to overcome key thermodynamic barriers that limit the production of reduced products where energy is stored in the form of C-H, H-H, and N-H bonds. The two interrelated research Thrusts of the *BETCy* EFRC emphasize mechanisms of electron bifurcation in driving low potential oxidation-reduction reactions. The research of the *BETCy* EFRC builds on recent seminal discoveries in biology and provides the basis for attacking key knowledge gaps and expanding the knowledge base that is essential for realizing the true potential of bioenergy and bio-inspired catalysis as prominent components of the global energy production portfolio. We have assembled a strong team of investigators with complementary research interests and technical skills to accomplish the proposed interdisciplinary tasks. The work is having a profound scientific impact on understanding and predicting matter and energy at the atomic level and in generating a blueprint for efficient control of electron flow into energy products and chemicals.

The work is directly in line with and addresses 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* (which is the essence of electron bifurcation reactions). 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 BETCy EFRC will address its scientific aims using advanced tools and technical approaches in support of the DOE-BES Transformative Opportunities: (1) Mastering Hierarchical Architectures and Beyond Equilibrium Matter, (2) Beyond Ideal Materials and Systems: Understanding the Critical Roles of Heterogeneity, Interfaces, and Disorder, (3) Revolutionary Advances in Models, Mathematics, Algorithms, Data, and Computing, and (4) Exploiting Transformative Advances in Imaging Capabilities across Multiple Scales. Oxidoreductase enzymes couple electrochemical potential to drive chemical transformations using complex architectures that integrate electron/proton circuits to enable the flow of matter over wide-ranging spatial and temporal scales. The complexity of scale presents an enormous technical barrier to elucidating functional principles of enzymes and exploiting their designs to realize transformative reactions. To address this critical issue, the BETCy EFRC team has developed experimental and theoretical methods at the forefront of redox reaction dynamics that enable measurements and interpretation of electron flow through enzyme circuits on the timescales of individual reaction steps.

Biological Electron Transfer and Catalysis Center (BETCy)	
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University of Georgia	Michael W.W. Adams (Associate Director)
Duke University	David Beratan
Montana State University	Brian Bothner
University of Washington	Caroline Harwood
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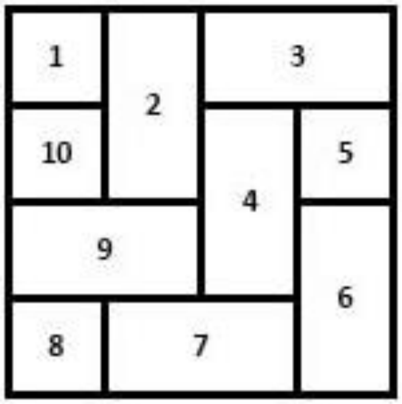
GRAND CHALLENGES INDEX

TRANSFORMATIVE OPPORTUNITIES INDEX

BES REPORTS INDEX

TOPICAL INDEX

EXPERIMENTAL AND THEORETICAL METHODS INDEX



FRONT COVER IMAGE

For more information about the images, see the corresponding EFRC technical summaries.

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2. The Center for the Science of Synthesis Across Scales (CSSAS) EFRC led by François Baneyx at the University of Washington (page 89)
3. Center for Lignocellulose Structure and Formation (CLSF) EFRC led by Daniel Cosgrove at the Pennsylvania State University (page 51)
4. Alliance for Molecular PhotoElectrode Design for Solar Fuels (AMPED) EFRC led by Gerald Meyer at the University of North Carolina at Chapel Hill (page 81)
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8. Center for Alkaline-Based Energy Solutions (CABES) EFRC led by Héctor Abruña at Cornell University (page 15)
9. Institute for Quantum Matter (IQM) EFRC led by Collin Broholm at Johns Hopkins University (page 25)
10. Center for Electrochemical Energy Science (CEES) EFRC led by Paul Fenter at Argonne National Laboratory (page 5)

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