Energy Frontier Research Centers

Technical Summaries

U.S. DEPARTMENT OF **ENERGY** | Office of Science
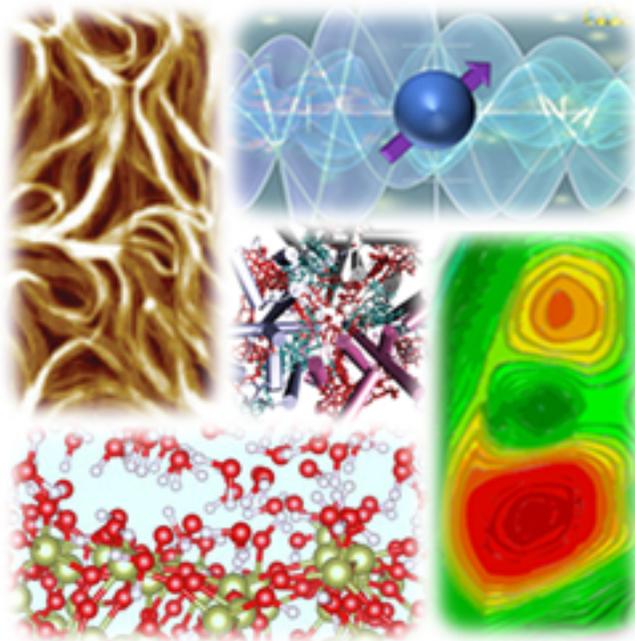
INTRODUCTION

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. 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. A targeted competition in environmental management, microelectronics, polymer upcycling, and quantum information science in 2020 resulted in 10 awards: 2 two-year extensions of existing EFRCs, 2 four-year renewals of existing EFRCs, and 6 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 41 EFRCs that are active in 2020 – 2022. 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 discover and understand new magnetic topological materials that host quantum phenomena and functionality for future applications in computing, spin-based electronics, 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. 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 (CATS) aim to discover and understand the properties of *magnetic* topological materials, including; magnetic TSMs, as well as 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 are developing 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 are being 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 is advancing our understanding and accelerating breakthrough innovations in TSMs. 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 for 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 discovers archetypal TSM compounds and delivers 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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Institute for Cooperative Upcycling of Plastics (iCOUP)
EFRC Director: Aaron Sadow
Lead Institution: Ames Laboratory
Class: 2020 – 2024

Mission Statement: To uncover macromolecular and catalytic phenomena at the interface of molecular-scale chemistry and mesoscale materials science to enable upcycling of energy-rich plastics.

The iCOUP research team is investigating the catalytic conversions of hydrocarbon polymers into more valuable chemicals and materials. Plastics are essential in the global economy, as reflected by production of new polymers in 2019 surpassing 400 million tons that consumed the equivalent of 6–8% of the crude oil and natural gas produced worldwide. Almost half of the currently manufactured plastics are polyolefins (POs), including polyethylene (PE), polypropylene (PP) and polystyrene (PS), 80% of which are single-use products discarded into overflowing landfills, contributing to a global waste catastrophe with widespread environmental, economic, and health-related consequences. Polyolefin upcycling requires the ability to break inert bonds in long chains of chemically indistinguishable repeat units at regular spatial intervals, thereby converting waste into targeted, narrow distributions of molecules and materials with desirable properties and added value.

Inspired by nature's approaches to biopolymer deconstruction, we are creating abiotic multifunctional materials that target and cleave specific bonds in macromolecules through three mechanistic motifs shown in **Figure 1**. The first strategy will achieve selective cleavage of carbon-carbon bonds in POs via molecular-scale mechanisms (**Figure 1A**), while those in **Figure 1B,C** will emphasize processive and polymer-site directed mechanisms akin to those in enzyme-catalyzed conversions. The iCOUP team will utilize their best features to create new upcycling methods which enable precise cleavages in polymer chains.
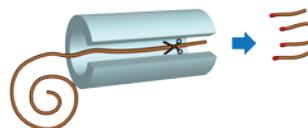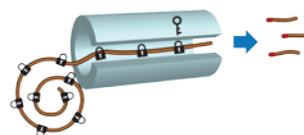
Figure 1. Three mechanistic motifs for polymer upcycling.

We will study how mesoscale architectures adapt conversions of small molecules, through these mechanistic motifs, to be useful for the upcycling of macromolecules. This scientific challenge will be addressed through studies focused upon catalytic sites, architectures, and polymers during deconstruction reactions in the following Objectives:

i. Discover new methods to transform intractable plastics into upcycled intermediates by breaking and functionalizing C–C and C–H bonds with molecular-scale selectivity;
ii. Design processive approaches to deconstruct polymers, leveraging molecular and macromolecular phenomena, to produce uniform, higher-value small molecules; and
iii. Construct next-generation POs containing sequences that facilitate end-of-life conversion.

The current state-of-the-art syntheses of heterogeneous catalysts revolve around the immediate molecular scale environment of reactive binding sites through generation of single sites, uniform nanoparticles, or ordered materials. The next and more daunting challenge is to create effective and selective catalytic architectures, operating at multiple length scales. The design and assembly of such

multifunctional catalytic systems that favor selective upcycling pathways relevant to specific polymers will require advances in synthesis, theory, and spectroscopy with the following Objectives:

    iv. Investigate synthetic and analytical methods for constructing and characterizing hierarchically-structured catalysts with spatially organized functional groups;

    v. Develop population balance and microkinetic models to relate experimental signatures of polymer deconstruction with macromolecular upcycling mechanisms; and

    vi. Predict how molecular-scale interactions and polymer conformational entropy govern the adsorption and mobility of polymers on surfaces (shown in Figure 2) and in mesoscale pores of catalysts.

By establishing the fundamental macromolecular phenomena germane to upcycling, our interdisciplinary team will create robust, selective inorganic catalysts and next-generation polymers that can be purposefully deconstructed and transformed into valuable, upcycled products. In a broader perspective, iCOUP's scientific advances create opportunities to depart from the current make-then-discard approach toward plastics and achieve a truly circular economy for these energy-rich resources.
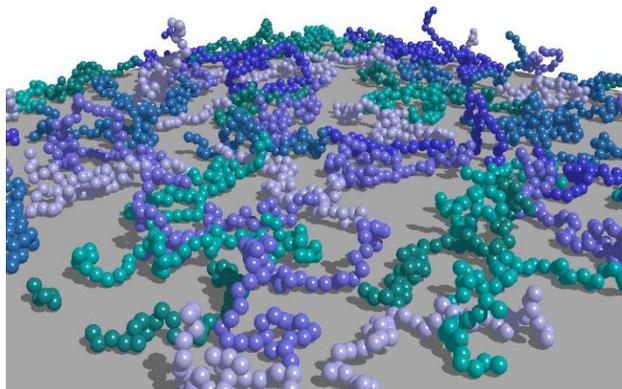
Figure 2. Typical conformation of polymers at a uniformly attractive surface, below the adsorption transition temperature.

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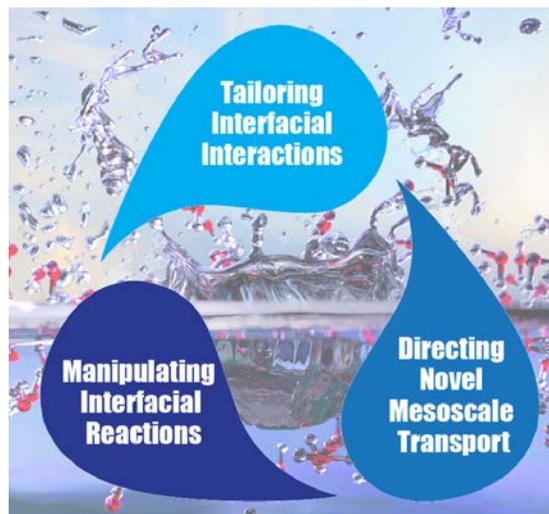
The AMEWS team brings together a confluence of capabilities to tackle the knowledge gaps outlined above. We have identified three integrated 4-year goals toward which we will work collectively as a center:

- 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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Ultra Materials for a Resilient, Smart Electricity Grid (ULTRA)
EFRC Director: Robert J. Nemanich
Lead Institution: Arizona State University
Class: 2020 – 2024

Mission Statement*: To achieve extreme electrical properties and phenomena through fundamental understanding of ultra wide bandgap materials – including synthesis and impurity incorporation, electronic structure at interfaces, electron - phonon interactions at high fields, and phonon mediated thermal transport, which will enable a resilient, smart electricity grid.*

A resilient, smart electricity grid is necessary to integrate multiple energy sources, power storage capabilities, and diverse electrical needs, and Ultra wide bandgap (UWBG) semiconductors have been identified as a crucial enabling materials technology. The UWBG semiconductor and dielectric materials (or 'Ultra' materials) present a new realm for high field transport, electron-phonon interactions, and heat transport. Understanding their novel properties will enable "reinventing the electricity grid" by providing efficient energy conversion and control (Smart Grid) and a significant reduction in size where a substation could be replaced by a suitcase-sized power converter (Resilient Grid).
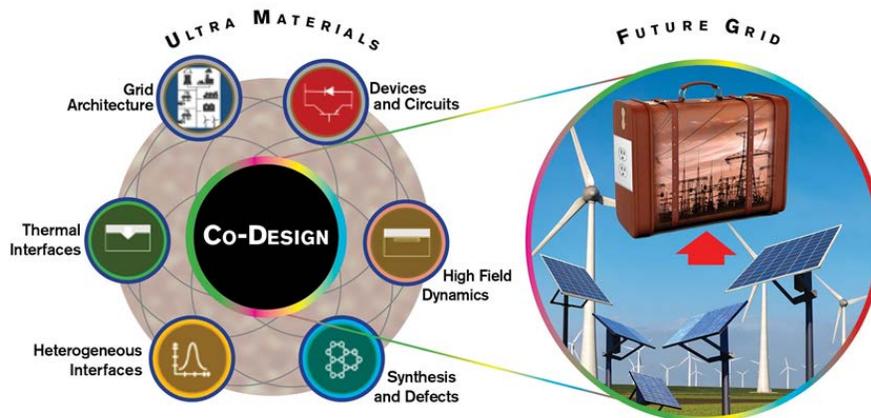
Figure 1. A Future Grid Co-Design Ecosystem will be established to enable communication across all levels of the science and technology.

The Mission of the Ultra EFRC is to understand fundamental phenomena in UWBG materials – including synthesis, defect and impurity incorporation, electronic structure at interfaces, interaction of electrons and atomic vibrations at high fields, to achieve extreme electrical properties, and efficient thermal transport. The Center will establish a co-design ecosystem enabling communication across all levels of the science and technology. The Center will focus on basic science challenges in four Thrusts: 1) growth, defects, and impurities, 2) heterogeneous interfaces, 3) carrier dynamics and high field transport, and 4) thermal energy transport and interfaces. The Ultra semiconductor materials of interest include cubic crystalline diamond, hexagonal crystalline AlN and the $B_xAl_{1-x}N$ alloy system which bridges the cubic and hexagonal crystal structure. The Ultra dielectric materials include oxide and fluoride thin films. The team brings together experts in non-equilibrium growth techniques, advanced microscopy, defect analysis, interface electronic states characterization, high field current transport, thermal properties, and thermal imaging measurements; this expertise is integrated with theory and modeling through a computational team that use *ab initio* first principles modeling, non-linear dynamics, self-consistent Monte Carlo heat transport, and high throughput simulations and materials informatics.

The research into new doping and interface configurations will be guided by high performance computing. As an example (Fig. 2), a multi-tiered computational screening approach will be used to identify low-energy, shallow-donor and acceptor defect configurations. The steps include importing the crystal structure, structural relaxation, high resolution simulation, high throughput simulations with correction terms included, computing formation energies, and selecting low formation energy shallow dopant configurations. Selected experimental results will be iterated into the process.

The Ultra EFRC will establish a Future Grid Co-Design Ecosystem, and develop a knowledge-base of UWBG materials and properties to "Reinvent the Electricity Grid." The outcomes will include: 1) synthesis of cubic and hexagonal UWBG semiconductors, 2) experimental and theoretical understanding of defects and doping that transcends the different materials systems, 3) characterized UWBG heterostructures enabling new routes to doping that exploit the properties of interfaces, 4) development of a deep understanding of electric breakdown phenomena and high current transport in UWBG semiconductors, and 5) characterized interactions between electrons and atomic vibrations and understanding the heat transport in UWBG materials and importantly, their interfaces. The research will provide a roadmap projecting how to achieve high breakdown field in the off-state, high current densities in the on-state, and highly efficient thermal conduction to minimize heating. The Future Grid Co-Design Ecosystem, will provide design simulation tools for a new generation of high power devices and power conversion modules and work with grid architect researchers to incorporate UWBG semiconductors in a Resilient, Smart Electricity Grid.

Figure 2. A high-throughput computational approach for identification of low formation energy and shallow donor and acceptor dopants in UWBG's.

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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.

Aim 2: Kinetics of interfacial reactions leading to corrosion. *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.
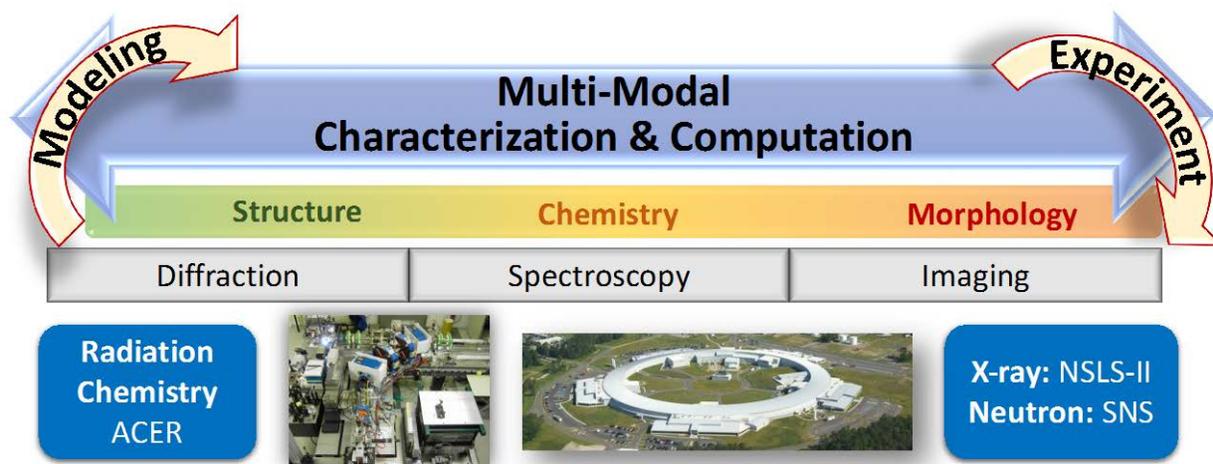
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Breakthrough Electrolytes for Energy Storage (BEES)
EFRC Director: Robert Savinell
Lead Institution: Case Western Reserve University
Class: 2018 – 2022

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.
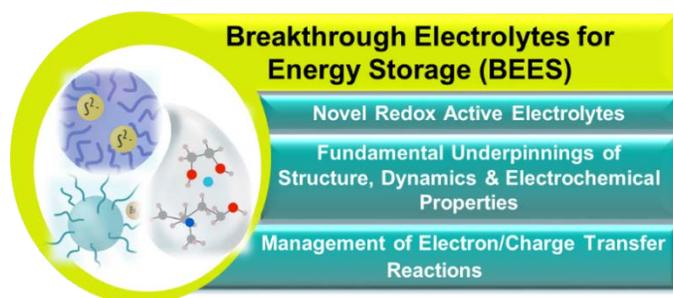
Figure 1. Research overview of BEES

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. The goal of Thrust 1 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 hypothesis of Thrust 1 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)	
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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

Synthesis & Characterization

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

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, Xavier Roy, P. James Shuck, Xiaoyang Zhu
University of Washington	Jiun-Haw Chu, David H. Cobden, Daniel Gamelin, Xiaodong Xu, Matthew Yankowitz
Carnegie Mellon University	Di Xiao

Contact: D. N. Basov, Pro-QM Director, db3056@columbia.edu
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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.

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).

(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 ca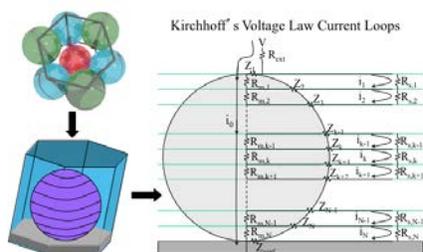talyst supports in alkaline media. We will evaluate new hierarchical motifs to harness their complex functionality. We will take advantage of recent findings that Nb-doped rutile TiO_2 supports retain their electronic conductivity, even under the strongly oxidizing electrochemical conditions of a fuel 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).
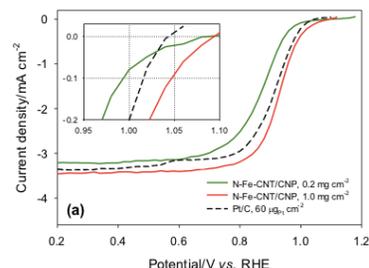
Fig. 1: ORR performance of N-Fe-CNT/CNP composite and Pt/C catalysts. Main Panel: RDE polarization plots in 0.1 M NaOH at 25°C and at 900 rpm. Inset: expanded view of the low overpotential region.

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.

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*-cyc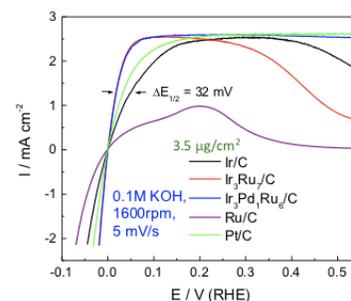looctenes and consequently, these functionalized building blocks can be polymerized in a living fashion. We will also seek to understand the mechanism of membrane degradation and potential carbonate formation and precipitation, as they will guide the structural design and optimization of ionomers with improved stability and high intrinsic conductivity.

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.

(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.

(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

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

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.

Summary: Over the four-year period, we foresee the CABES effort as providing the basis for ushering in an Alkaline-Based Energy Technology Society.
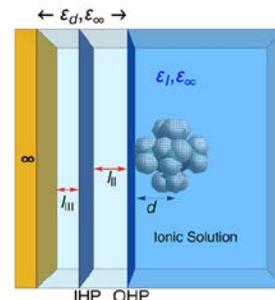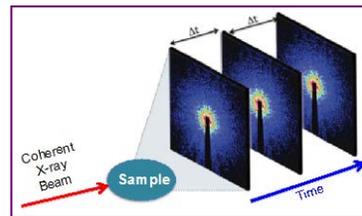
Center for Alkaline Based Energy Solutions (CABES)	
Cornell University	Héctor Abruña (Director), Tomas Arias (Thrust Area Leader), Geoffrey Coates (Thrust Area Leader), David Muller (Thrust Area Leader), Joel Brock, Peng Chen, Francis DiSalvo, Robert DiStasio, Emmanuel Giannelis, Lena Kourkoutis, Andrej Singer, Jin Suntivich, Paul Mutolo (Executive Director)
University of Pennsylvania	Tom Mallouk (Thrust Area Leader)
Binghamton University	Jiye Fang
University of Wisconsin	Manos Mavrikakis
Carnegie Melon University	Kevin Noonan
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National Renewable Energy Lab	Bryan Pivovar
Yale University	Sharon Hammes-Schiffer

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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-Schoenzart (Director), Susan Latturner, Kenneth Hanson, Eugene DePrince, Joseph Sperling
Florida International University	Ines Triay (Deputy Director), Christopher Dares
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
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**Center for Understanding and Control of Acid Gas-Induced
Evolution of Materials for Energy (UNCAGE-ME)
EFRC Director: Ryan P. Lively
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.
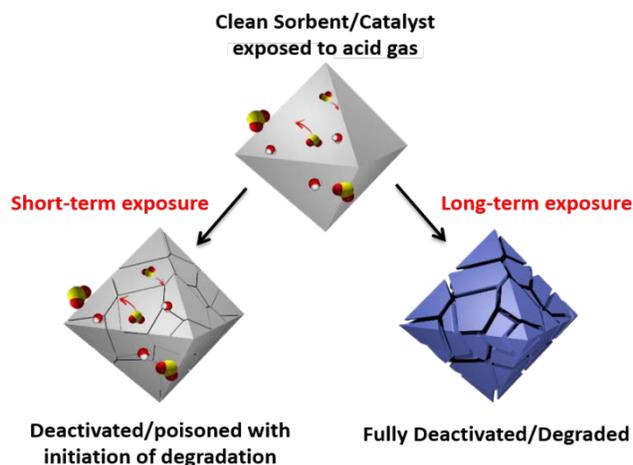

Figure 1: Changes induced in sorbents/catalysts upon short- and long-term exposure to acid gases. Materials are typically treated as passive in these environments, but can change dramatically upon exposure to SO_x, NO_x, and H_2S.

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.
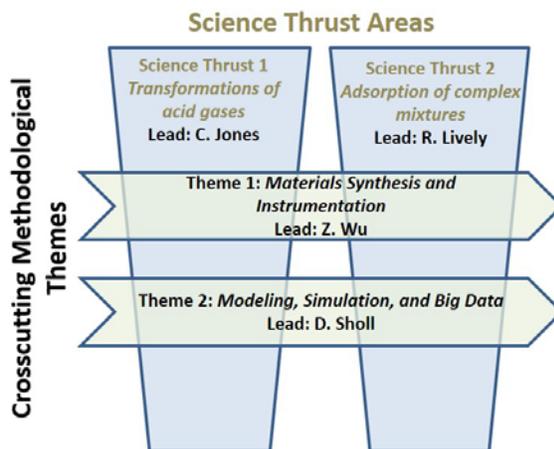
An overview of Science Thrusts and cross cutting Methodological Themes for Phase II of UNCAGE-ME is illustrated in Fig. 2. These Thrusts and Themes will work in an interconnected manner. The Science Thrusts focus on two broad strategies for developing the fundamental science for understanding and control 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.

Figure 2: Overview of research thrusts/themes.

Center for Understanding and Control of Acid Gas-Induced Evolution of Materials for Energy (UNCAGE-ME)	
Georgia Institute of Technology	Ryan Lively (Director), Christopher Jones, David Sholl, Sankar Nair, Rampi Ramprasad, Krista Walton
Oak Ridge National Laboratory	Katharine Page, Bobby Sumpter, Zili Wu
Sandia National Laboratories	Jessica Rimsza, 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

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

Mission Statement: Improve catalytic selectivity by quantitatively scaling from model studies to catalytic conditions using advanced experiment and theory with a strategic objective to develop overarching design principles.

The vision of IMASC is to advance the fundamental science necessary to reduce the carbon footprint of the chemical industries sector. Specifically, the mission is to develop the ability to increase energy efficiency through improvement of catalytic selectivity using dilute alloy catalysts, by quantitatively scaling from model studies to catalytic conditions using advanced experiment and theory. The principle governing the use of dilute alloy catalysts is that the reactive minority metal, e.g. Pd, Ni, Pt, and Ag, initiates the catalytic cycle, whereas the abundant host, Au or Ag, imparts selectivity. A strategic objective is to develop overarching design principles and to develop a general methodology for understanding catalytic processes.

Background, knowledge gaps, Mission and Goals

Chemical production, which relies heavily on heterogeneous catalysis, now accounts for nearly 25% of energy use worldwide. Forecasts for global energy demand project this number will rise to 45% by 2040. The Center's mission and operational imperative is rooted in a singular reality: current trends in energy supply and use are unsustainable—economically, environmentally, and socially. A fully integrated theoretical-computational-experimental approach to the design of selective catalysts is needed to boost energy efficiency in the industry's production processes. IMASC 2.0 answers this call, with significant progress already made at the intersection of surface chemistry and physics to transform how catalysts are designed.

Heterogeneous catalytic processes are extremely complex, requiring optimization of factors across multiple scales of length, time, pressure, and temperature. To develop such catalyst processes mandates team science and an interdisciplinary approach, inclusive of materials synthesis, mechanistic surface chemistry, reaction kinetics and *in-situ* and *operando* characterization. Numerous studies show that complex metal/oxide interfaces, generally present in catalysts, appear to play an important synergistic role in determining reactivity. Further, since materials are often affected by the reaction environment, pre- and on-stream activation and optimization of performance is necessary.

Historically, heterogeneous catalytic processes have been devised empirically, with broad guidelines informed by prior experience in organic and organometallic chemistry. Recent advances in theory and experiments provide tools with the potential to move beyond the traditional "trial-and-error" approach to design principles that predict and develop highly efficient heterogeneous catalysis materials systems. IMASC bridges this divide and address key knowledge gaps motivated by key basic research needs identified by DOE.

Four scientific goals support this mission

Design and synthesize robust nanoporous dilute alloy catalysts.

IMASC has developed a toolbox of dilute, alloy catalyst materials designed to provide insight into catalyst function. Two classes, a free-standing nanoporous material and a raspberry colloid templated nanoparticle material, each demonstrate excellent properties. They exhibit remarkably stable catalytic performance and impart a high degree of reaction selectivity.

Predict catalytic selectivity through understanding of kinetics and mechanism.

A central goal of IMASC is to predict reaction selectivity under catalytic conditions based on reaction mechanisms and kinetics determined from fundamental studies. In IMASC remarkable progress has been made on the research objective to advance the paradigm for control of selective oxidation and hydrogenation on dilute alloy catalysts and to quantitatively model their kinetics.

Exploit rearrangement at interfaces to enhance selectivity and activity.

IMASC is focusing on a critical challenge in heterogeneous catalysis—the development of catalytic processes that achieve and maintain high activity and high selectivity through optimization of the composition and structure of dilute alloy catalysts. A key principle underlying our approach is to devise an approach whereby the minority more reactive metal is at or near the surface so it can initiate the catalytic cycle; e.g. O_2 activation for oxidation or H_2 dissociation for hydrogenation

Advance methodology for catalytic design.

Understanding the evolution of structure and composition of catalysts and their relationship to catalytic function is a central goal of IMASC that is enabled by the development and implementation of advanced characterization and theoretical modeling. Experimental tools that have been advanced in IMASC include *in operando* atomic-scale imaging using electron microscopy, structural determination using EXAFS in combination with machine learning, and atom probe tomography for 3-D imaging of composition using atom probe tomography. Theoretical tools have also been advanced in IMASC research that model catalyst restructuring using accelerated molecular dynamics, and kinetic modeling of reactive steps by including a more accurate treatment of pre-exponential factors.

Integrated Mesoscale Architectures for Sustainable Catalysis (IMASC)	
Harvard University	Joanna Aizenberg, Cynthia Friend (Director), Boris Kozinsky, Robert Madix
LLNL	Juergen Biener
BNL	J Anibal Boscoboinik, Dario Stacchiola
Stony Brook University	Anatoly Frenkel
Tulane University	Matthew Montemore
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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 $5f$ 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 $5f$-electron materials:

1. What is the impact of $5f$ 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 $5f$ 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.
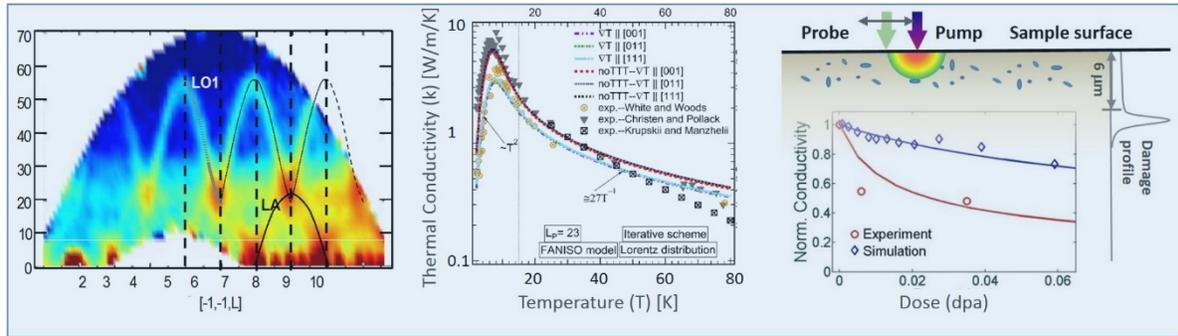
Figure. 1. *Left*: Dynamic structure of UO$_2$ measured along Q-[-1,-1,L] at 400k using the ARCS instrument at the Spallation Neutron Source, Oak Ridge National Laboratory. *Middle*: Simulation of the thermal conductivity of solid argon using the Boltzmann Transport Equation approach. *Right*: Time-resolved 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$_x$O$_2$ 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)	
Idaho National Laboratory	David Hurley (Director), Jian Gan (Deputy Director), Mukesh Bachhav, Michael Benson, Krzysztof Gofryk, Lingfeng He, Zilong Hua, Chao Jiang
Oak Ridge National Laboratory	Michael Manley
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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.
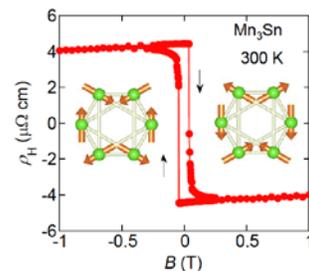

Fig. 1: Hall resistivity at room *T* in Mn_3Sn with spin configurations.

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.
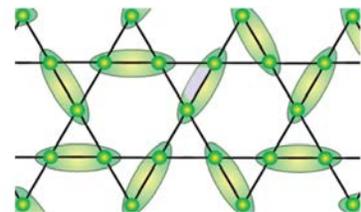

Fig. 2: A valence-bond configuration on a 2-dimensional kagome lattice. In a quantum spin liquid valence bonds resonate.

Topology in Superconductivity. When a superconductor is cooled below Tc, an energy gap opens in the single particle density of states, forming a gap much like that found in an insulator. Consequently, the recent advances demonstrating the crucial importance of topology, and the incompleteness of prior classification schemes, in true insulators should apply to superconductors as well. Recent work by IQM has shown that the traditional superconductor classification scheme, based on spherical harmonics, is incomplete, missing those that require "monopole" harmonics (Fig. 3). IQM will design, synthesize, and characterize materials and nano-scale structures to achieve and document a physical realization of monopole superconductivity.
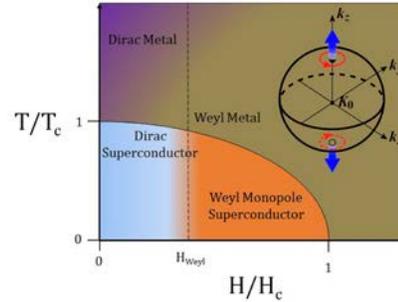
Fig. 3: The monopole superconductor has vorticity and may be realized in a Weyl semimetal.

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.
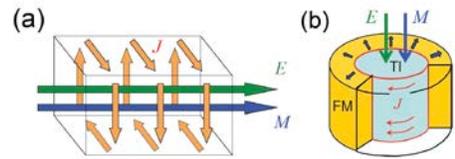

Fig. 4: Connection between magneto-electric coupling and surface current (a). TIs and axion insulators have a quantized surface anomalous hall effect. For TIs the surface states must be gapped by a ferromagnetic cladding layer (b) but this occurs spontaneously in axion insulators.

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.

Institute for Quantum Matter (IQM)	
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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.

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
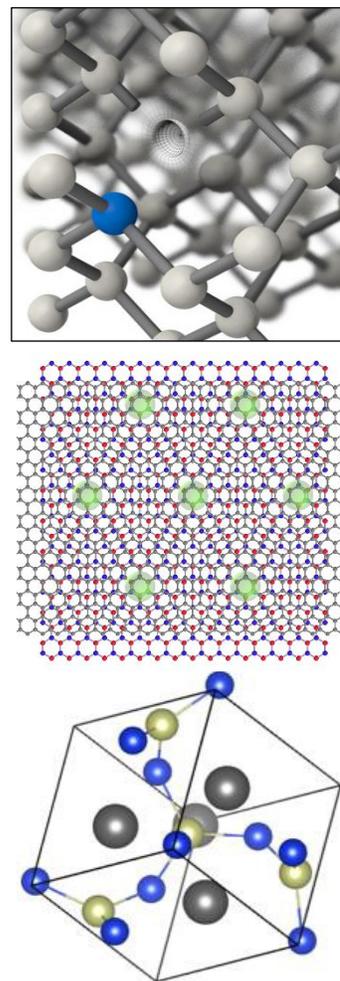

Figure 1. Illustrations of research directions for the three thrusts of this EFRC. *From top to bottom*: a nitrogen-vacancy (NV) center in the diamond lattice used for quantum sensing; 2D layers of trilayer graphene and boron nitride form a superconducting moiré superlattice; crystal structure of LaIrSi,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

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.

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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.
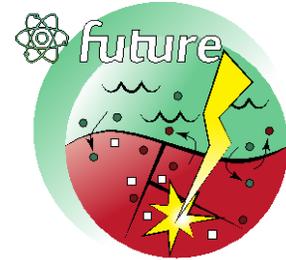
In FUTURE, we target the response of materials to a combined corrosive and irradiation environment at relevant conditions of temperature and stress. Corrosion is driven by mass transport to and from reactive surfaces, across interfaces, and/or through protective scale layers. At the same time, the transport 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 are developing unique experimental capabilities that target these questions. In the case of point defect generation during irradiation, we complement in situ transmission electron microscopy (TEM) studies with 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 are designing a PAS capability on LANL's existing ion beam lines to quantify these defects in situ as irradiation is happening, as opposed to typical studies in which defect content is quantified after the fact.

These studies are complemented by advanced microscopy characterization that examines how defects couple with elemental species in the material to modify overall evolution. In particular, we use advanced

4D microscopy techniques to examine how microstructural features interact with defects and alloying elements to modify transport pathways. Uniquely, we are using isotopic atom probe tomography (APT) to determine how species such as oxygen migrate through the microstructure. By using isotopic labels, we can determine those regions in which transport is enhanced most significantly.

Finally, we are targeting transport across the liquid/solid interface. We are using electrochemical impedance spectroscopy (EIS) to quantify key reaction rates at these interfaces. We are also expanding on a unique capability in which a material is exposed simultaneously to a coupled irradiation and corrosion environment. This irradiation-corrosion experiment (ICE) has been used to study liquid metal corrosion. We are expanding this capability 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.
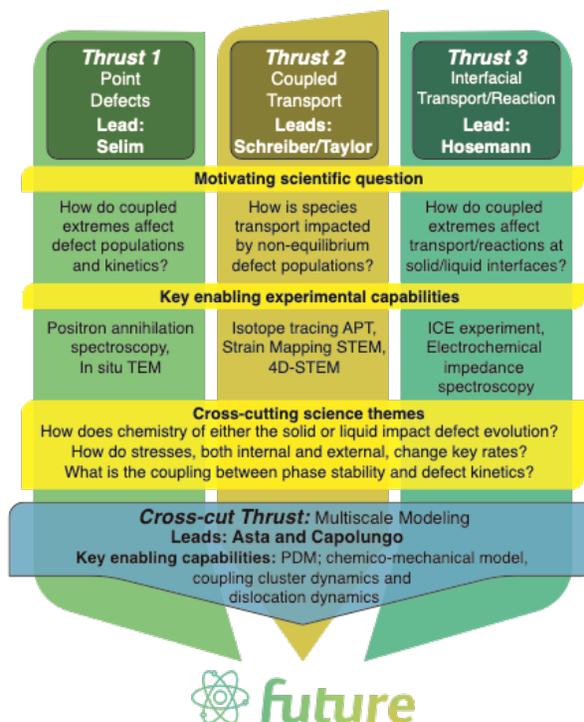
All of this experimental activity is complemented by a cross-cutting modeling activity that aims to both elucidate the mechanisms observed in the experiments and develop a capability to predict the evolution of materials under these coupled extremes. The heart of this multi-scale effort is a new capability 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. This modeling framework enables 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.
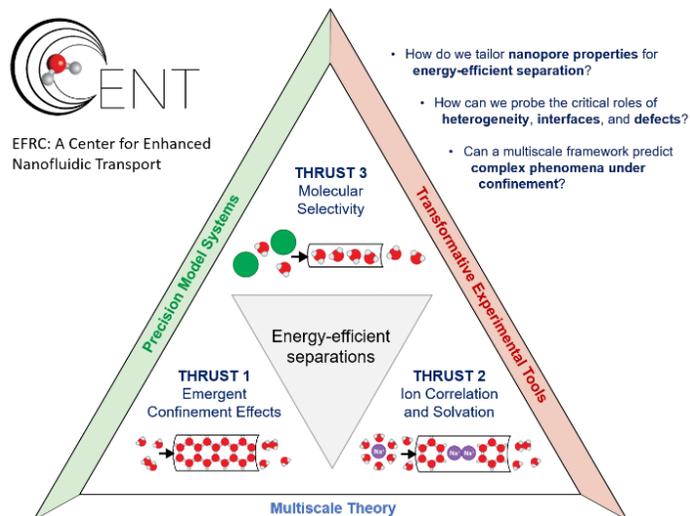
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Center for Enhanced Nanofluidic Transport (CENT)
EFRC Director: Michael S. Strano
Lead Institution: Massachusetts Institute of Technology
Class: 2018 – 2022

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.

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-

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

Gibbs-Thomson phase behavior – that fluid phase boundaries in SDNs are remarkably distorted from their bulk fluid counterparts; and highly non-linear, correlative effects in ion transport through SDNs 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.
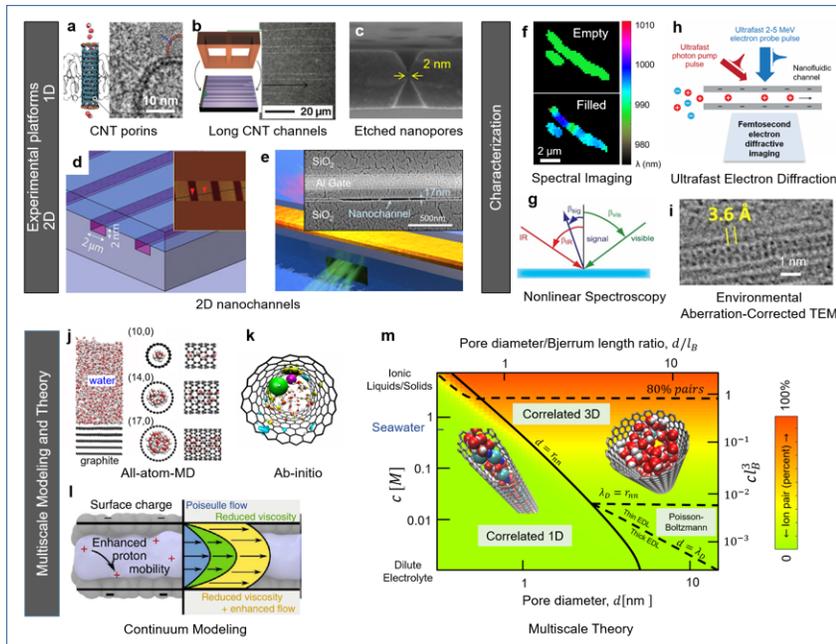
The Center for Enhanced Nanofluidic Transport (CENT) addresses 7 key nanofluidics knowledge gaps: (1) Slip Flow Enhancement in SDNs; (2) Fluid Phase Transitions in SDNs; (3) Phase-Separation under Confinement; (4) Defects and their Outsized Impact on SDN Transport; (5) Correlated Transport; (6) Nanoscale Solvation; and (7) Charge Exclusion to Significantly Enhance Selectivity. The 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 are addressing solvation effects in aqueous media and in acetonitrile,

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 also explore fundamentally new mechanisms of ionic and molecular selectivity that stem from extreme confinement. CENT is pioneering 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 set the scientific basis for

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.

developing next generation membrane materials. CENT also expands the ability to incorporate controlled, single defects into SDNs as a novel perturbation method and explores the impact of pore defects on nanofluidic transport. The CENT team pioneers 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 is producing 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.

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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$_3$NH$_3^+$), 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.

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,
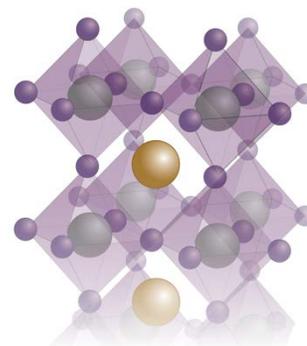

Figure 1(a) ABX$_3$. A (gold sphere) site is the organic component, B site is the center of the octahedra, X site are tips of the octahedra.
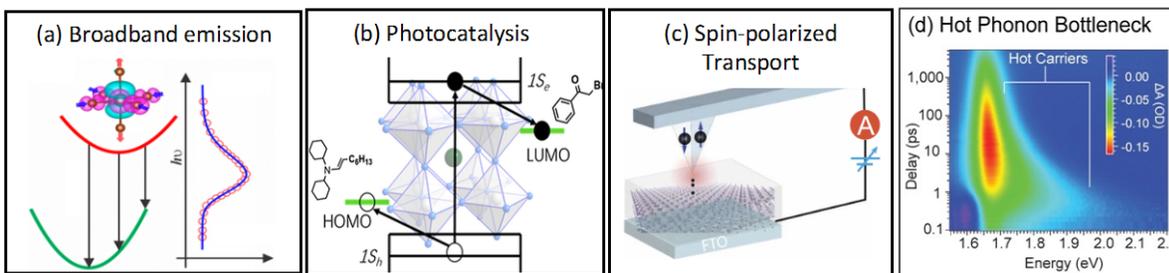
Figure 2. (a) Broadband emission. (b) Photocatalysis.(c) Spin transport in Chiral systems (d) A hot phonon bottleneck dramatically slows carrier cooling.

significantly influencing the structural, electronic, optical, and defect properties, and leading to emergent properties (Fig.2) that include: dynamic behavior, ferroelasticity, ferroelectricity, 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 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.

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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 Molecular Quantum Transduction (CMQT)
EFRC Director: Michael R. Wasielewski
Lead Institution: Northwestern University
Class: 2020 – 2024

Mission Statement: To develop the fundamental scientific understanding needed to carry out quantum-to-quantum transduction through a bottom-up synthetic approach, which imparts atomistic precision to quantum systems.

Quantum-to-quantum transduction is the coherent exchange of information between quantum systems, which is an essential element of quantum information science. To achieve this goal, **CMQT** explores the underlying interactions among quantum spins, excitons, and vibrational excitations of molecules and molecular materials that are relevant to molecular quantum-to-quantum transduction. **CMQT** comprises an interdisciplinary team of chemists, physicists, and materials scientists with the individual expertise and collective breadth to create knowledge in this emerging area.

Why use molecules and why now? To date, research in spin-based QIS has demonstrated success by harnessing and exploiting defects in solids. Using molecule-based systems offers the advantages of structural reproducibility, atomic scale spatial control, structural modularity, and access to uniquely molecular degrees of freedom (DOFs), i.e. the various pairwise interactions between photons, excitons, magnons, phonons, spins, and charges (**Fig. 1**). The number of quantum DOFs available in molecular systems make them attractive targets for quantum transduction, as quantum information can be transferred coherently between DOFs.
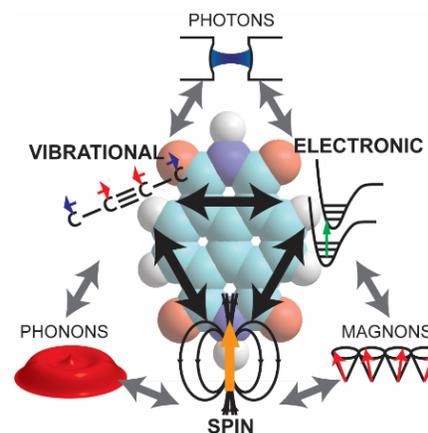
Fig. 1. Synergistic **CMQT** research directions. Thrust 1 focuses mainly on spin and electronic DOFs. Thrust 2 adds phononic and magnonic DOFs. Thrust 3 adds photonic and vibrational DOFs.

Molecular architectures provide unmatched flexibility for tailoring the properties that are critical to quantum transduction, and molecular synthesis affords the opportunity to build novel molecular materials from the bottom-up, both of which are at the heart of our proposed research. Thus, molecular systems offer an exceptional opportunity to explore the interface between quantum systems essential for sensing, communication, and computation.

CMQT exploits recent breakthroughs from our team including landmark coherence times and stabilities of molecular qubits and quantum materials, the ability to create hybrid qubits, and resonant photonic architectures. As we move forward, our approach includes both ensemble-level studies to rapidly understand interactions, and development of *single-molecule* methods to interface molecular QIS with other QIS platforms. We are also leveraging cutting-edge physical measurement techniques with high spatial, temporal, and spectral resolution to understand how to transition quantum-to-quantum transduction from the ensemble to the single molecule level.

Our goals are embodied by three cross-cutting Thrusts with closely integrated approaches and team synergies that progressively exploit the flexibility and tunability of molecular architectures to address quantum-to-quantum transduction at increasing length scales. Individually, the Thrusts each pose and answer fundamental questions relevant to quantum transduction in different regimes, ranging from local to long-distance. Taken together, the Thrusts develop a transformative integrated framework for how

molecules can facilitate quantum transduction at all the scales relevant for quantum information science.

Thrust 1. Localized Molecular Quantum-to-Quantum Transduction (co-Leaders: Fuchs and Freedman). *The goal of this Thrust is to develop new mechanisms and strategies to coherently couple localized molecular DOFs and thus lay the foundation for molecular quantum-to-quantum transduction.* Designer molecular qubits with long coherence times and tunable interactions enable quantum state transduction between molecular quantum states demonstrated at the ensemble level to be transitioned rapidly to quantum measurement at the single molecule level. We are exploring synthesis and measurements that leverage atomic precision to enable quantum transduction through local interactions.

Thrust 2. Distributed Molecular Quantum-to-Quantum Transduction (co-Leaders: Johnston-Halperin and Long). *The goal of this Thrust is to demonstrate quantum transduction within distributed molecular quantum systems.* Thrust 2 explores quantum transduction in ensembles of tailored molecular qubits including those developed in Thrust 1 that interact *via* spin-magnon coupling to delocalized, highly coherent, magnon modes in molecule-based magnetic thin films. This approach bridges the length scales of single molecules with those of state-of-the-art solid-state quantum systems.

Thrust 3. Multiscale Molecular Quantum-to-Quantum Transduction (co-Leaders: Goldsmith and Weiss). *The goal of this Thrust is to use the combination of flying qubits (photons) and molecular DOFs to achieve quantum transduction over multiple length scales within hierarchical quantum systems.* Thrust 3 incorporates the molecular systems established in Thrusts 1 and 2 into photonic structures to demonstrate coherence transfer between multiple molecular DOFs and between these DOFs and photons, including producing heralded photons necessary to probe quantum aspects of energy-important light-harvesting processes, such as natural and artificial photosynthesis.

Achieving molecular quantum-to-quantum transduction is necessarily an interdisciplinary effort, requiring the scope of an EFRC to assemble the necessary expertise in the design and synthesis of molecular and solid-state materials, the capacity to measure coherent quantum states at the single quantum level, and the ability to seamlessly incorporate theory and modeling of materials and measurement schemes with the experimental constraints of real systems. **CMQT** is working toward meeting this challenge.

Center for Molecular Quantum Transduction (CMQT)	
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Princeton University	Nathalie de Leon
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University of Iowa	Michael Flatté
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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 develops 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 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:
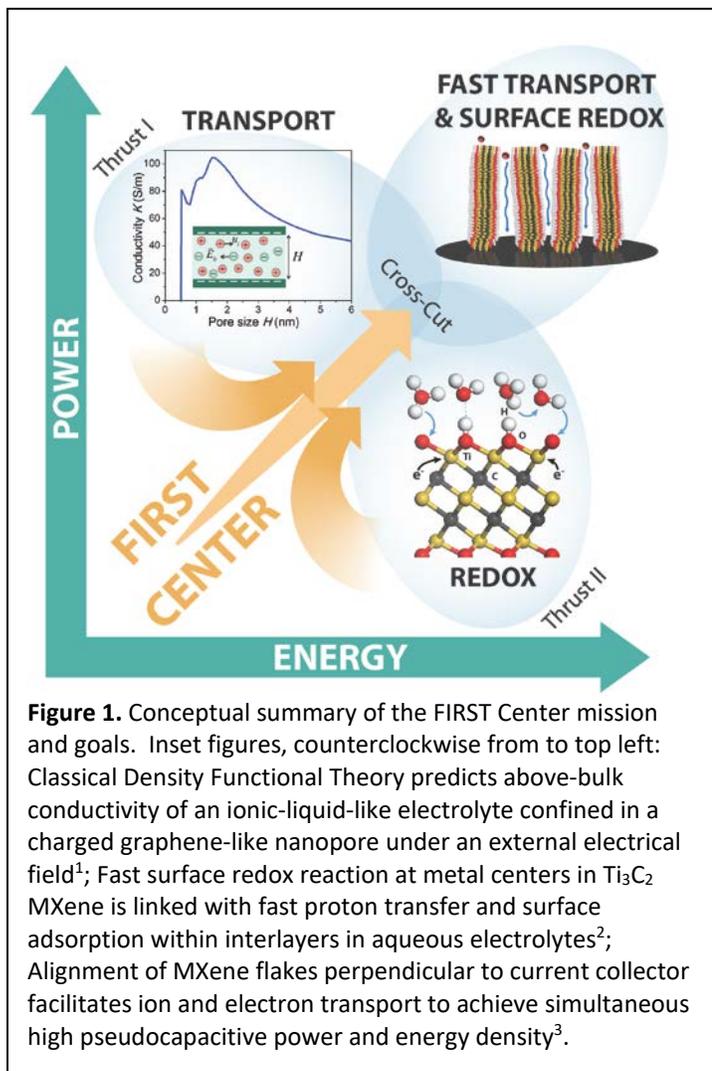
Figure 1. Conceptual summary of the FIRST Center mission and goals. Inset figures, counterclockwise from to top left: Classical Density Functional Theory predicts above-bulk conductivity of an ionic-liquid-like electrolyte confined in a charged graphene-like nanopore under an external electrical field[1]; 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.

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[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).

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[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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North Carolina State University	Veronica Augustyn
Penn State University	Adri Van Duin
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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 – 2022

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.
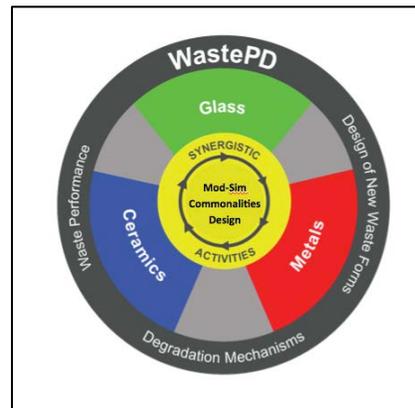
Crystalline ceramic waste forms can also release the incorporated radionuclides during interaction with the environment through matrix dissolution, selective leaching, radionuclide diffusion and surface reaction. An alteration phase may also form on the surface of ceramic waste forms to protect against 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.*
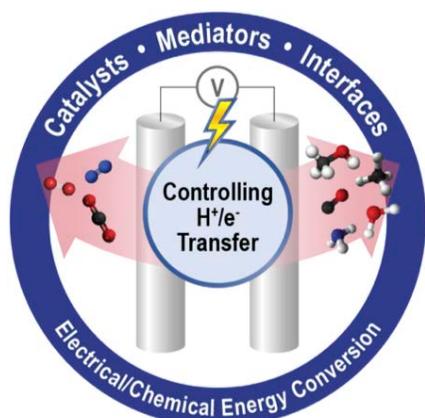

Figure 1. Efficient electrocatalysts require precise control of movement of protons and electrons.

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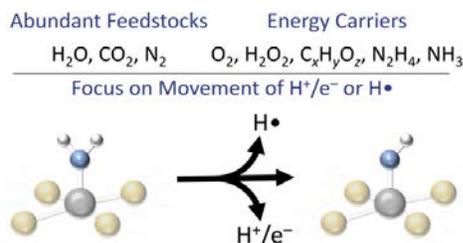
Figure 3. By focusing on the thermodynamics and kinetics of the movement of protons/electrons, CME develops the principles needed to design catalysts with unprecedented performance.

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 are showing how the mechanistic characterization of catalytic reactions reveals not one, but multiple scaling relationships. Our work contributes 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
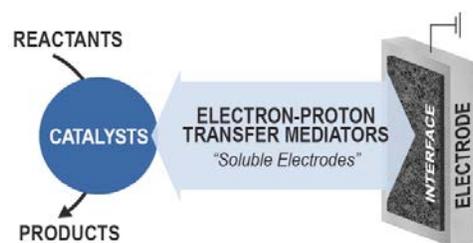
Figure 4. Mediated catalyst design is an emerging concept that improves system performance by delivering H^+/e^- away from the electrode.

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 addresses 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 guide the exploration and optimization of the complex free energy landscape of catalytic processes. Methodological developments 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 CME research 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.
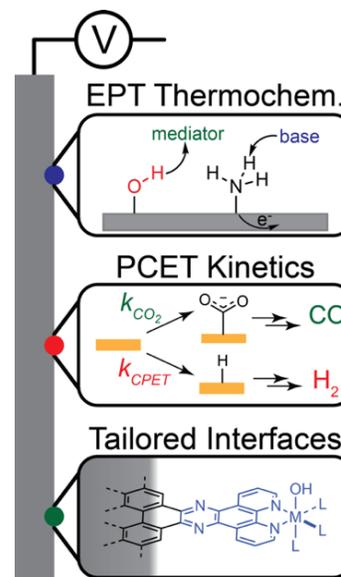
Center for Molecular Electrocatalysis (CME)	
Pacific Northwest National Laboratory	Morris Bullock (Director), Aaron Appel (Lead, Catalysis Design Thrust), Simone Raugei (Lead, Theory/ Computations), Eric Wiedner
Yale University	Sharon Hammes-Schiffer (Deputy Director), James Mayer (Lead, Heterogeneous Interfaces Thrust)
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Interfacial Dynamics in Radioactive Environments and Materials (IDREAM)
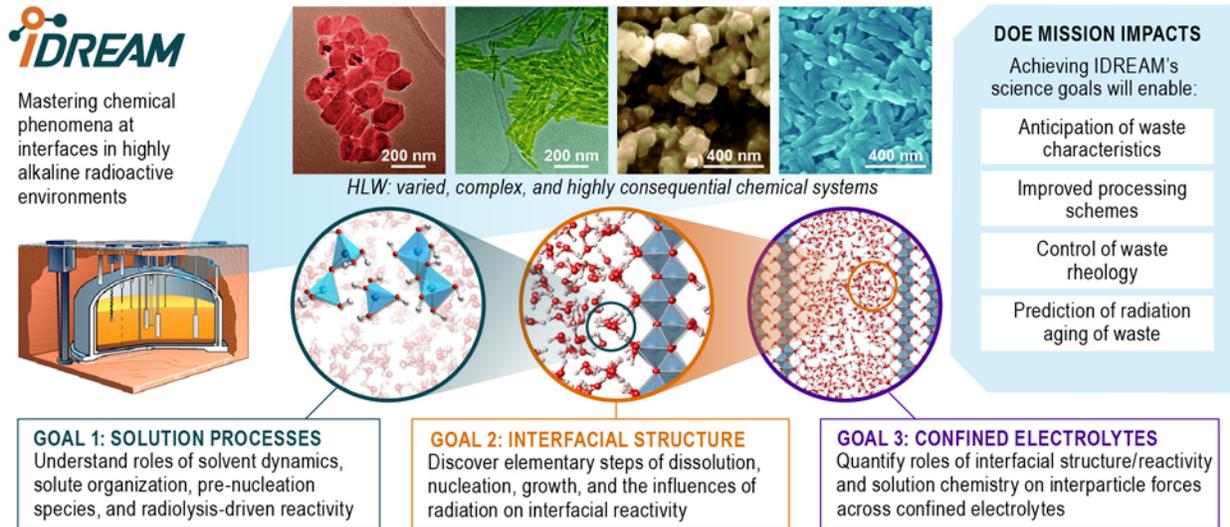**EFRC Director: Sue Clark**
Lead Institution: Pacific Northwest National Laboratory
Class: 2016 – 2024

Mission Statement: To master interfacial chemistry in complex environments characterized by extremes in alkalinity and low-water activity, where molecular phenomena are driven far from equilibrium by ionizing radiation.

IDREAM is revealing the chemical driving forces for ion behavior in complex alkaline electrolytes at interfaces exposed to ionizing radiation. Experimental and computational studies are integrated to discover the roles of ion networks, long-range solvent structure, and steady-state transient species in solution and interfacial reactivity. IDREAM achieves this mission by pursuing research organized with the following Science Thrusts:

1. Science Thrust 1 (**ST1**) on Molecular and Solution Processes: *Understand solvent dynamics, solute organization, prenucleation species, and radiation-driven reactivity in concentrated alkaline electrolytes.*

2. Science Thrust 2 (**ST2**) on Interfacial Structure and Reactivity: *Discover the elementary steps of dissolution, nucleation, and growth, and the influence of radiation on interfacial reactivity in highly alkaline systems of concentrated electrolytes.*

3. Science Thrust 3 (**ST3**) on the Dynamics of Confined Electrolytes: *Understand the chemical and radiation-driven phenomena of nanoscale confined electrolytes that lead to interactions between interfaces to form aggregates of particles.*

DOE MISSION IMPACTS
Achieving IDREAM's science goals will enable:
- Anticipation of waste characteristics
- Improved processing schemes
- Control of waste rheology
- Prediction of radiation aging of waste

HLW: varied, complex, and highly consequential chemical systems

Mastering chemical phenomena at interfaces in highly alkaline radioactive environments

GOAL 1: SOLUTION PROCESSES
Understand roles of solvent dynamics, solute organization, pre-nucleation species, and radiolysis-driven reactivity

GOAL 2: INTERFACIAL STRUCTURE
Discover elementary steps of dissolution, nucleation, growth, and the influences of radiation on interfacial reactivity

GOAL 3: CONFINED ELECTROLYTES
Quantify roles of interfacial structure/reactivity and solution chemistry on interparticle forces across confined electrolytes

IDREAM is providing the fundamental science basis to process the millions of gallons of highly radioactive wastes stored at DOE's Hanford and Savannah River Sites. With currently available technologies, removing these wastes from tanks and stabilizing them for disposal will take decades and will cost hundreds of billions of dollars. Building on IDREAM's research progress, our research goals advance a foundation of use-inspired knowledge, enabling accelerated waste-processing alternatives.

IDREAM accelerates discovery through a cross-cutting theme that reveals the role of very rapid radiolysis processes in generating steady-state transient species. These species drive the nonequilibrium chemical dynamics, and their role in solution speciation and formation of ion clusters in not well understood. Their presence also impacts nucleation, growth, and dissolution of solids in these complex alkaline electrolytes.

IDREAM is also developing new theories of reaction dynamics and interfacial chemistry. IDREAM employs unified and novel experimental, computational, and theoretical approaches that take full advantage of Office of Science experimental and computational user facilities. Our experimental efforts will involve multimodal spectroscopic approaches, including neutron scattering, X-ray total scattering/pair distribution function (TS/PDF) analysis, quasi-elastic neutron scattering, synchrotron-based X-ray absorption spectroscopies, NMR, time-resolved X-ray spectroscopies, and vibrational spectroscopies (e.g., Raman and infrared). Our experimental approach is closely integrated with our computational efforts, providing a theoretical basis for the observed spectroscopic signatures and extending our efforts to extract unique spectroscopic contributions from species ensembles using sub-ensemble analysis. Finally, IDREAM is producing well-defined materials through controlled synthesis, including radiation-driven synthesis. IDREAM is advancing revolutionary new approaches to enable control of matter driven far from equilibrium by radiation, particularly at interfaces.

The primary mission impact is Environmental Management. The U.S. Department of Energy (DOE) faces significant challenges with high-level waste (HLW) storage, retrieval, and processing and currently estimates that completion of HLW cleanup will require at least 50 years and hundreds of billions of dollars. Many issues limit current HLW processing schedules, including costly characterization needs, slow treatment processes, and unpredictable rheology of waste streams. The new knowledge gained from IDREAM will provide the scientific foundation for innovations in characterization and processing strategies. These innovations will serve DOE and the American public by accelerating HLW treatment and significantly reducing the aggregate costs for HLW cleanup.

Interfacial Dynamics in Radioactive Environments and Materials (IDREAM)	
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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.
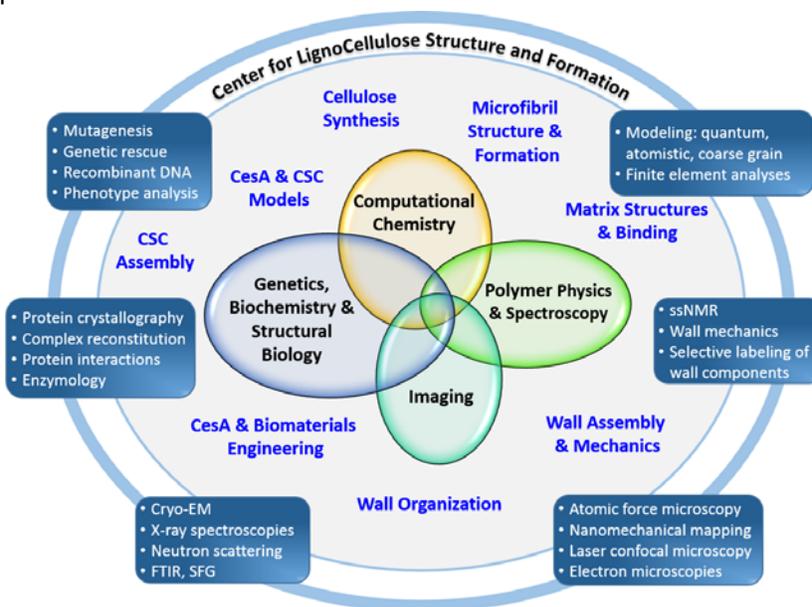
CLSF goals in the current phase will build upon advances made in the previous funding period to:

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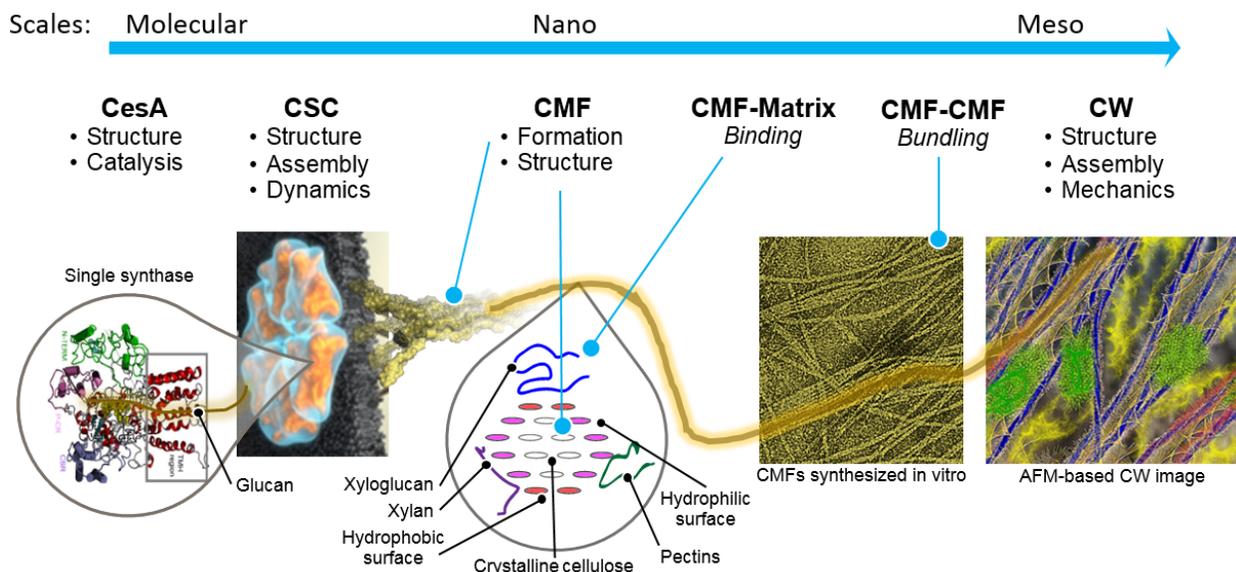
Above: Research questions addressed by CLSF include the structure and kinetics of cellulose synthase (CesA); the structure and activity of the cellulose synthesis complex (CSC); cellulose microfibril (CMF) structure and CMF interactions with water, matrix polysaccharides and lignin.

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
University of Virginia	Jochen Zimmer
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Three-Dimensional Ferroelectric Microelectronics (3DFeM)
EFRC Director: Susan Trolier-McKinstry
Lead Institution: The Pennsylvania State University
Class: 2020 – 2024

Mission Statement*: To exploit the 3rd dimension in microelectronics for functions beyond interconnects by incorporating non-volatile ferroelectric memory densely interconnected with logic to create low-power, 3D non-von Neumann computation.*

The *Center for 3D Ferroelectric Microelectronics (3DFeM)* will explore the 3rd dimension in microelectronics for *functions beyond interconnects,* enabling 3D **non-von Neumann** *computer architectures exploiting ferroelectrics for local memory, logic in memory, digital/analog computation, and neuromorphic functionality*. This approach circumvents the end of Moore's law in 2D scaling, while simultaneously overcoming the "von Neumann bottleneck" in moving instructions and data between separate logic and memory circuits. Empowered by new ferroelectric materials that overcome 60-year-old materials compatibility challenges, *3DFeM will tackle the non-von Neumann challenge* to propel radical advances in microelectronic devices, circuits, and systems. **3DFeM will enable a million-fold enhancement in interconnection between memory and logic, along with substantial reductions in the energy cost to computation**.
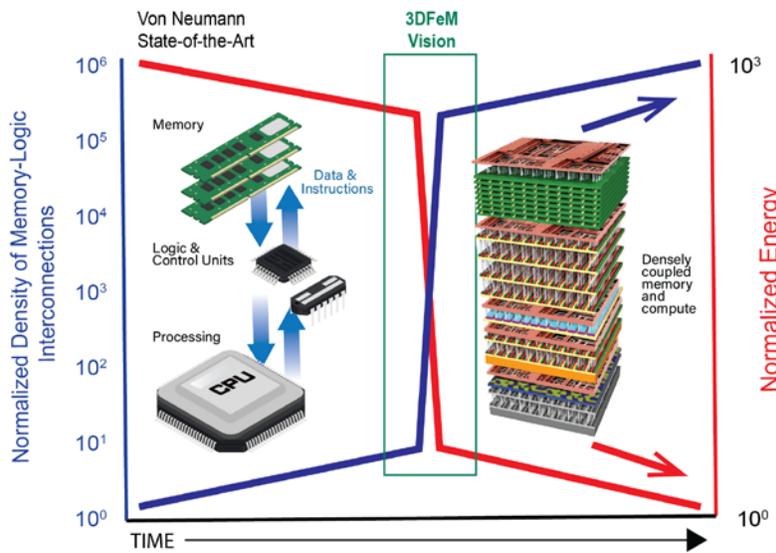
Figure 1: Existing computer architectures are predominantly based on the von Neumann architectural paradigm, where the memory and computation logic are fundamentally separable components. Thus, time and energy are lost in communicating between separate chips. The 3DFEM vision breaks the von Neumann bottleneck by integrating ferroelectric memory and computing elements in 3D, so the circuit interconnects become functional. 3DFeM would enable radical increases in memory-logic connectivity.

Computing accounts for 5 – 15% of worldwide energy consumption. While recent efficiency gains in hardware have partially mitigated the rising energy consumption of computing, major gains are achievable in a paradigm shift to 3D computing systems. The 3DFeM program will enable novel energy efficient hardware (Fig. 1). In many applications, ferroelectrics provide a 10^3-10^4 × improvement in energy efficiency relative to other technologies, while also reducing latency and area overhead. In addition, ferroelectrics enable new functionality for power management, embedded intelligence, and security for the Internet of Things (IoT), impacting a wide range of industries spanning transportation to defense to residential building.

However, there are bottlenecks associated with fundamental understanding of ferroelectric physics and processing.

> **3DFeM Guiding Questions**
> - How can ferroelectricity be obtained in 3D-integration-enabling materials?
> - How can materials be made far from equilibrium (an essential component of BEOL integration) using synthesis routes consistent with 3D integration and aggressive scaling?
> - How can new devices leverage the cooperative nature of ferroelectricity, and can the cooperative nature of ferroelectricity be tuned for specific device requirements?
> - Can new properties, such as ferroelectric quantum critical points; superconductivity; and spin, charge, or orbital ordering, enable novel devices?
> - How can intrinsic or engineered properties of ferroelectric materials and devices enable novel compute-in-memory processors?

By closely coupling experiments, modeling, and theory, 3DFeM is pursuing foundational advances in materials, and devices by composing novel ferroelectric-based morphable compute elements, by answering the challenging scientific questions shown above (See 3DFeM Guiding Questions box). 3DFeM leverages world-leading expertise in integrated ferroelectrics, advanced characterization, and novel devices. Advanced Micro Devices, Applied Materials, IBM, Intel, GlobalFoundries, and Kurt J. Lesker Company are engaged 3DFeM partners.

3DFeM will achieve these goals through two strongly interconnected research thrusts. Thrust 1 will explore the basic mechanisms underpinning ferroelectricity and switching pathways. The thrust will focus on understanding ferroelectricity in HfO_2 and AlN-based systems; quantifying their properties and performance expectations; finding new BEOL-compatible ferroelectric phases; and developing the scientific underpinnings for ferroelectricity in new materials to guide their discovery and future use.
Thrust 2 will develop the fabrication processes needed for fabrication of ferroelectric devices and associated science and technology to validate Thrust 1 materials developments Thrusts 1 and 2 are linked by synchronized and continuous intersection to ensure: (1) nascent ferroelectric formulations are evaluated for compatibility with BEOL integration, (2) device design leverages realistic properties, and (3) emergent synthesis tools expand capability as it relates to both device performance and integration.

Three-Dimensional Ferroelectric Microelectronics (3DFeM)	
The Pennsylvania State University	Susan Trolier-McKinstry (Director), Vijaykrishnan Narayanan (Associate Director), Jon-Paul Maria (Thrust 1 leader), Thomas N. Jackson (Thrust 2 leader), Nasim Alem, Ismaila Dabo, Roman Engel-Herbert, Venkat Gopalan, Qi Li, Ying Liu, Shashank Priya
Purdue University	Shriram Ramanathan
RIT	Kai Ni
University of Virginia	Jon F. Ihlefeld
University of Pennsylvania	Andrew M. Rappe
Oak Ridge National Laboratory	Sergei Kalinin, Nina Balke-Wissinger, Stephen Jesse
Sandia National Laboratories	Thomas Beechem, Giovanni Esteves, Michael David Henry, Sean Smith

Contact: Susan Trolier-McKinstry, 3DFeM Director, STMcKinstry@psu.edu
814-863-8348

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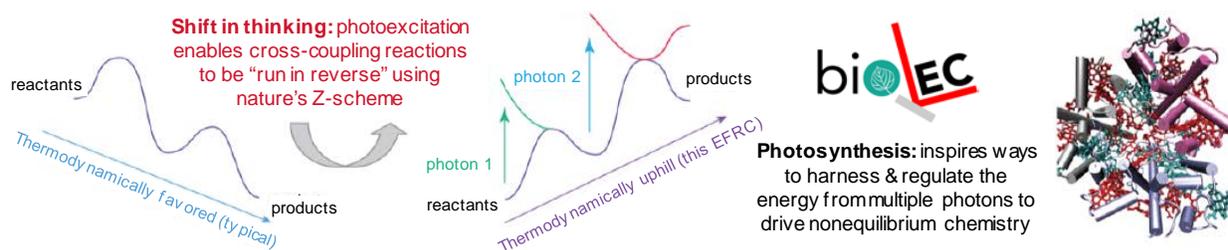
Figure 1. BioLEC's interdisciplinary approach will enable nonequilibrium chemistry.

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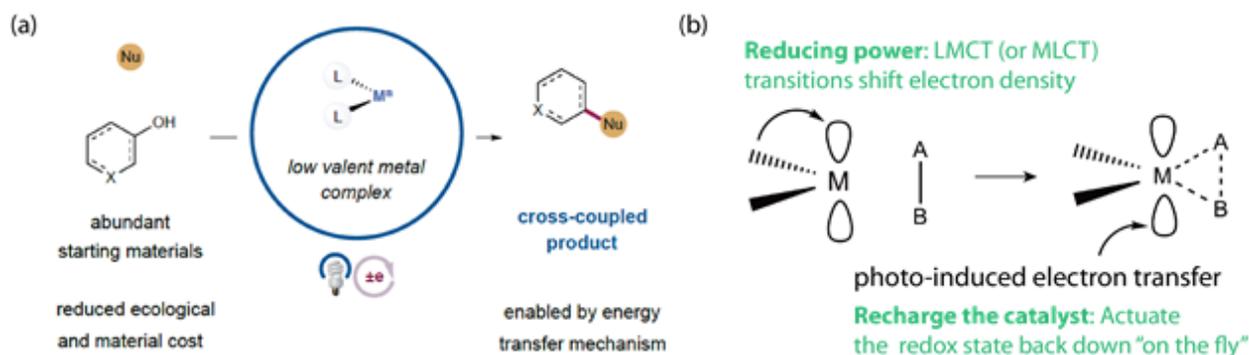
Figure 2. (a) Activation of inert bonds for cross-coupling. (b) BioLEC will step-change the limits of equilibrium redox potentials by adding or removing electrons "on the fly," thereby driving reactions forward.

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
NREL / University of Colorado, Boulder	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.

- **Research Group 1 – Materials Design and Discovery:** Using novel quantum-electro-dynamic and first-principles calculations, the RG1 team is identifying conditions that maximize the useful work of a photonic system, involving energy, phase, momenta, and entropy of photons. In parallel, using state-of-the-art materials chemistry, we are
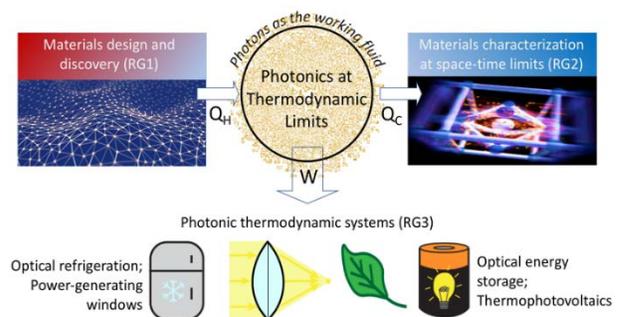
Figure 1: Schematic overview of our "Photonics at Thermodynamic Limits" Energy Frontier Research Center.

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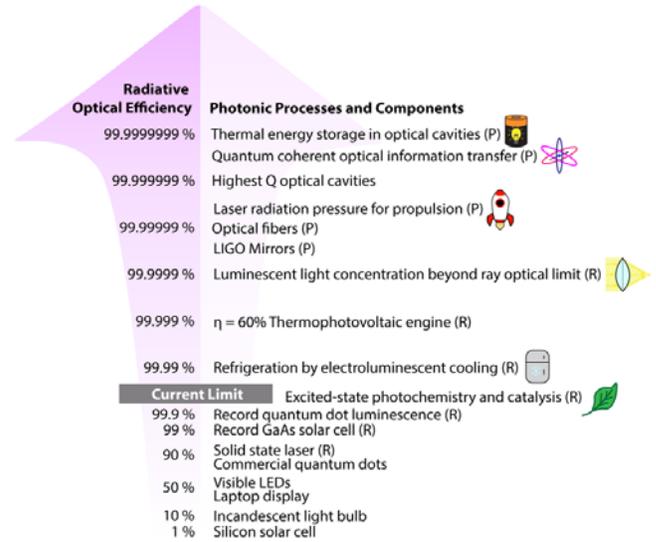
Figure 2: *Passive (P) and Active radiative (R) systems that become possible with increasingly high optical efficiency.*

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:

- Developed new excited state theoretical methods to describe electron/phonon and photon/phonon interactions in nanoparticles and 2D materials (RG1).
- Achieved record conversion efficiency in a regenerative thermophotovoltaic system, incorporating photonic design to reuse low-energy photons (RG3).
- Realized near-unity luminescence in core/shell quantum dots (RG1) and characterized their quantum yield with high-precision using photothermal threshold quantum yield measurements (RG2).
- Designed novel heterostructures of two-dimensional transition metal dichalcogenides (RG1).
- Correlated optical and electron microscopy to identify the role of structure on quantum emission in two-dimensional materials (RG2).
- Utilized ultrafast electron diffraction (UED) at SLAC to unravel nanocrystal structural dynamics (RG2).
- Developed new paths to non-reciprocal emission and transmission (RG3).

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.*

CMC-UF features a tightly integrated fundamental research effort combining experimental, theoretical, and numerical science. We are motivated by the economic, strategic, and environmental importance of unconventional formations, i.e., shale resources, and the lack of mechanistic understanding of coupled transport, reaction, and mechanical processes in such natural nanoporous media interlaced with fractures, Fig. 1. Fundamental physical and chemical understanding is foundational to achieve first-order improvements in environmental outcomes and recovery.
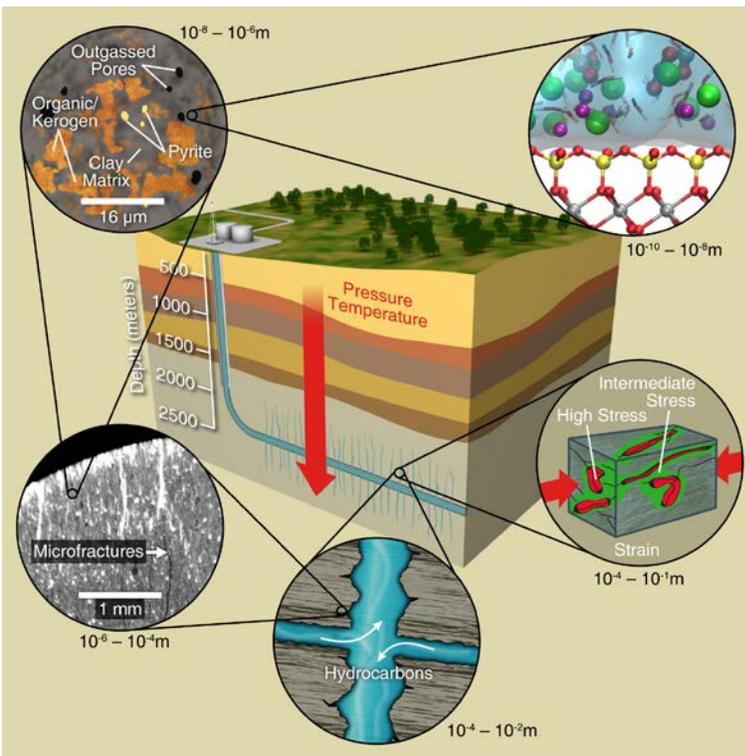
Fig. 1. Overlapping length scales of shale features of interest to CMC-UF.
From the upper right: water/clay interactions in a 10 nm wide slit pore where water is shaded blue; nanoCT image of shale structure illustrating minerals, kerogen, and outgassed pores; micro-fractures filled with barite scale; matrix-to-fracture mass transfer; zones of enhanced ductility in heterogeneous shale.

Our research approach is comprehensive and interdisciplinary. Importantly, five overarching research themes (i.e., discipline-specific expertise) are crosscut by seven multidisciplinary, synergistic research activities to foster acquisition of fundamental knowledge, as shown in Table 1. CMC-UF has made significant progress in unraveling the interplay of material heterogeneity, transport through heterogeneous structures, and fluid interactions with shale mineral/organic-matter interfaces within the context of a natural disordered system as a function of stress and transport process.

We are exploiting and creating new imaging and image reconstruction capabilities before, during, and after reaction to explore the largely inaccessible interior of the shale matrix. We employ X-ray computed tomography (CT), positron emission tomography (PET), scanning electron microscopy, scanning transmission electron microscopy and nuclear magnetic resonance (NMR) to achieve resolution from cm to nm length scales of reactive transport processes. We are pioneering chemical and physical description of reactive imbibition where rates of mass transport are of the same order as rates of chemical reaction. Importantly, we are developing and applying machine learning and data analytics to achieve image super resolution and to facilitate interpretation among measurement modalities.

Machine learning also features in our efforts to translate the impact of physical phenomena across scale. Convective and diffusive mass transfer is studied jointly with imaged dynamic experiments employing microfluidics, x-ray CT, and PET. Experiments are conducted in concert with at-scale numerical models including molecular dynamics, direct numerical simulation, lattice Boltzmann methods, and continuum formulations. Translation of the importance of physical mechanisms across length and time scales is a unifying activity within the research plan as indicated by the progression of scales in Fig. 1 and the activities in Table 1.

Table 1. Matrix of research expertise cross cutting the 7 major synergistic research activities in CMC-UF. All activities incorporate experimental measurements, theory development, and activities to translate results across length and time scales.

	Research expertise				
	Characterization methods	Transport processes	Reactivity at interfaces	Geomechanics	Translation
Synergistic activities	Mechanics of shale in the presence of reactive and nonreactive aqueous and nonaqueous fluids				
	Reactive imbibition of aqueous fluids in concert with evolution of tight matrix pore and microfracture structure				
	Reactive transport in fractured media from the pore to the continuum scale				
	Sorption and capillary condensation in nanoporous geomaterials				
	Phase transitions in tight materials in comparison to bulk properties				
	Probing advection and diffusion in nanoporous matrix from pore to pore network to core scale				
	Matrix-fracture interactions of tight matrix communicating with microfractures in the context of transport, reactions, and mechanics				

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 Illinois at Urbana Champaign	Jennifer Druhan
University of Wisconsin	Christopher Zahasky
University of Wyoming	Vladimir Alvarado, Saman Aryana, Teresa Lehmann
University of Southern California	Kristian Jessen, Theo T. Tsotsis
SLAC National Accelerator Laboratory	John Bargar, Gordon Brown

Contact: Anthony R. Kovscek, Director, kovscek@stanford.edu,
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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 actively control the pathways that govern synthesis and lead to new materials.*

Our ability to solve application-driven energy problems—such as the production, conversion and storage of energy, or the design of tailored heterogeneous catalysts and catalyst substrates—depends on the development of next-generation functional materials with targeted properties. While computational materials discovery can now predict new hypothetical materials (and their functional properties), computation has struggled to predict the inherently non-equilibrium processes that govern their synthesis. Realizing these hypothetical functional materials through conventional intuition-guided synthesis is a slow, rate-limiting, iterative process.
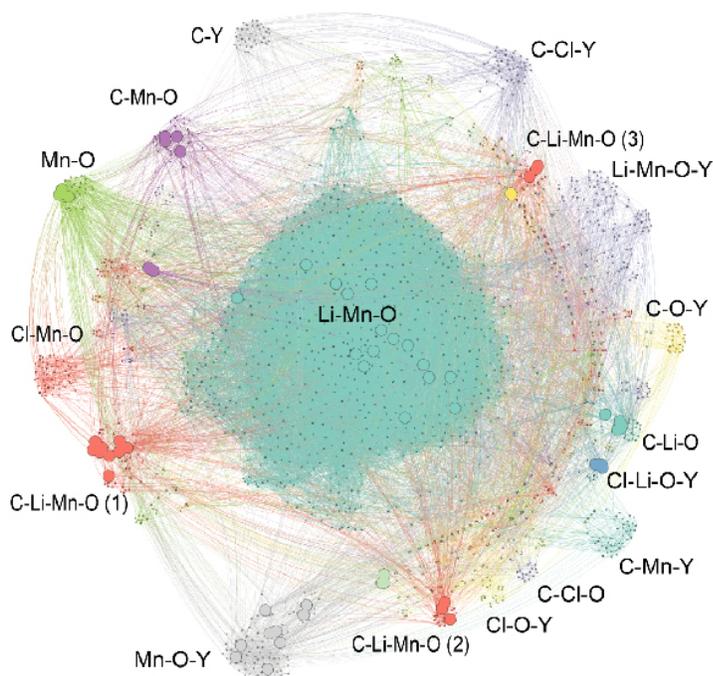
The reaction network for the C-Cl-Li-Mn-O-Y chemical system. The network contains 56 phases: 39 stable and 17 metastable. Chemical subsystems are labeled by color. The larger nodes indicate reactant nodes which are traversed on the 20 shortest pathways from precursors to targets in YMnO$_3$ synthesis.

To be *useful*, a material must be *made*, and its synthesis must then be optimized, regardless of how it was conceived or serendipitously discovered. Control over synthesis pathways and products is a critical requirement for materials design, without which concepts developed *in silico* cannot be brought into reality. Can we develop a data-driven approach to design synthesis pathways *ab initio?*

A radical rethinking of inorganic materials synthesis is needed; one that moves us forward from think-cook-look-repeat strategies to machine-predicted synthesis pathways, in a manner analogous to computational property-driven structure-prediction methods. Several emerging concepts in experimental and computational approaches, especially in combination, hint at ways forward.

Chemical reaction network models, constructed from thermochemistry databases, offer computationally tractable approaches for suggesting likely reaction pathways via application of pathfinding algorithms, presenting new opportunities for enabling reaction pathway prediction, rapid iteration between experimental/theoretical results, and ultimately, control of synthesis.

GENESIS workflow with in situ experimental probes analyzed in real time to enable controlled, next-generation synthesis.
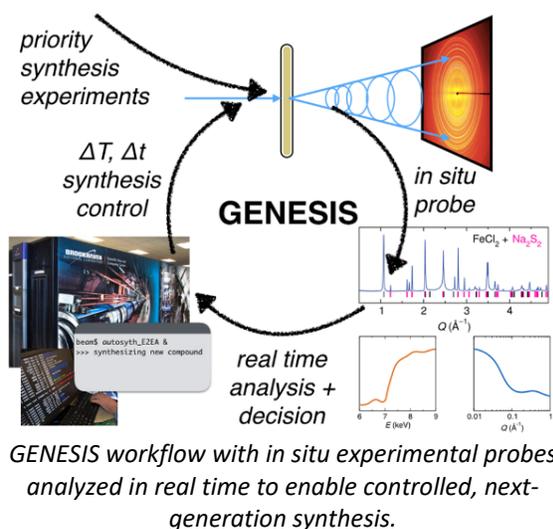
Bright sources of X-rays and neutrons at National Facilities allow users to perform high-throughput studies to determine local and long-range atomic arrangements, site chemistries, stress-strain relationships, sample textures in inorganic materials as a function of p, T etc. Access to open searchable databases and powerful computational resources lead to lists of target structures, prioritized by the fundamental properties leading to the desired functionality, and synthesizability. Development and deployment of individual experimental and computational tools are not transformative in themselves. Their combination and a focus on the reaction pathway, rather than just the beginning and end points of a reaction, will produce a transformation in synthesis science.

The GENESIS effort complements other DOE efforts. Specifically, the *de novo* prediction and adaptive control of synthesis routes enabled by GENESIS can facilitate the realization of new materials identified within programs, including hypothetical materials identified through computational materials discovery or materials that implement new design rules. The tools for science-based synthesis, including at large DOE user facilities, serve a broader community to understand and control the synthesis different families of materials beyond those of specific interest to GENESIS. The GENESIS framework for understanding how to effectively access materials away from equilibrium and, build new infrastructure to rapidly collect and interpret *in situ* synthesis data that can be use by any researcher in this field.

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	Eric Dooryhee
Columbia University	Simon Billinge
Colorado State University	James Neilson
Farmingdale State College	Jack Simonson
Lawrence Berkeley National Laboratory	Kristin Persson, Gerbrand Ceder
Oak Ridge National Laboratory	Gabriel Veith
University of California, San Diego	Ping Liu
University of Michigan	Katsuyo Thornton

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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 base necessary to enable future creation of scalable electrochemical energy storage systems with high energy, high 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 interfacial phenomena by deliberate design and manipulation of dynamic interfaces.

SG3. Gain fundamental understanding of design and function of thick porous electrode 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 electronic 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 composition, 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 from multiscale modeling, theory, and experiment, 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

Figure 1. Multiscale transport in electrode architectures.

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.

Center for Mesoscale Transport Properties (m2m/t)	
Stony Brook University	Esther Takeuchi (Director), Amy Marschilok (Deputy Director), Carlos Colosqui, 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
Georgia Institute of Technology	Tom Fuller
Kennesaw State University	Altug Poyraz
Lawrence Berkeley Laboratory	Marca Doeff
Lehigh University	Elsa Reichmanis
University of Texas at Austin	Guihua Yu

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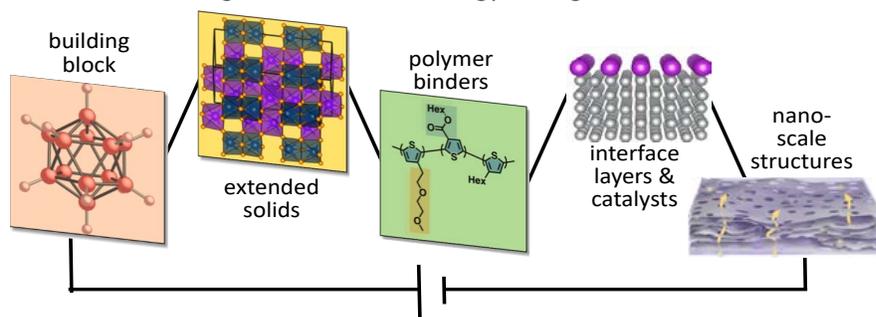
Figure 1: The SCALAR EFRC aims to rethink battery materials, starting from the level of building blocks and then moving to new motifs for extended solids. New materials are integrated with multifunctional polymer binders, designer interfaces, and controlled nanoscale architecture.

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

Contact: Sarah Tolbert, Director, tolbert@chem.ucla.edu
(310) 206-4767, www.chem.ucla.edu/SCALAR/

Quantum-Materials for Energy Efficient Neuromorphic- Computing (Q-MEEN-C)
EFRC Director: 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").*
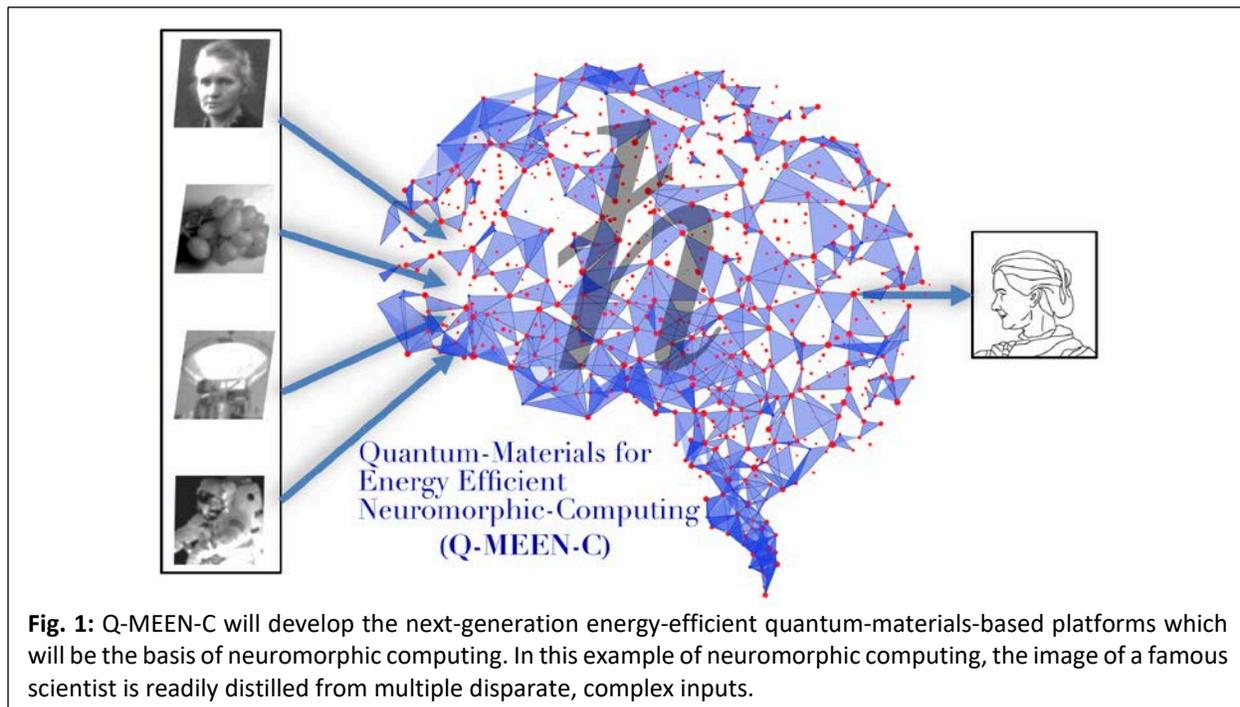
Fig. 1: Q-MEEN-C will develop the next-generation energy-efficient quantum-materials-based platforms which will be the basis of neuromorphic computing. In this example of neuromorphic computing, the image of a famous scientist is readily distilled from multiple disparate, complex inputs.

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.

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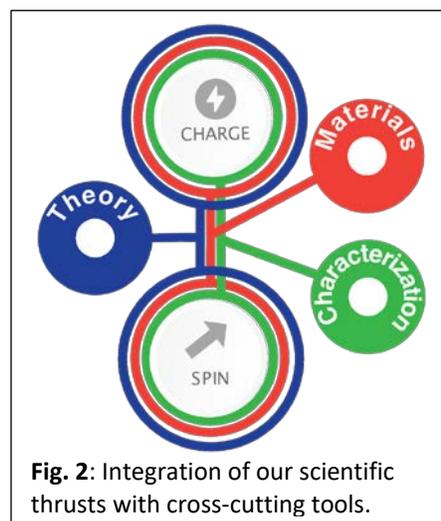
Fig. 2: Integration of our scientific thrusts with cross-cutting tools.

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
Northwestern University	Amanda Petford-Long
National Center for Scientific Research	Julie Groliere, Marcelo Rozenberg
University of Illinois, Urbana-Champaign	Axel Hoffman
NIST / University of Maryland	Mark Stiles

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Center for Plastics Innovation (CPI)
EFRC Director: LaShanda Korley
Lead Institution: University of Delaware
Class: 2020 – 2024

Mission Statement: To develop catalytic and functionalization approaches and fundamental tools applicable to the upcycling of polymer plastics waste, with a strategic focus on enabling mixed-stream transformations from varied material form factors.

We will develop a comprehensive polymer plastics waste (PPW) upcycling strategy that combines fundamental discoveries in catalytic technology and chemical functionalization with innovations in polymer design and additive manufacturing and is enabled by the leveraging of computational, data science, characterization, and systems design tools. We will target these scientific pursuits toward the transformation of complex PPW streams into high-value fuels, lubricants, monomers, and functional polymers. Specific to our approach is the integration of multiscale considerations of evolving materials complexity during upcycling, predictive strategies to accelerate discovery, and less energy-intensive

processing considerations. These objectives will nucleate a transition from the current high-energy and lower-value landscape of polymer recycling to a new frontier of polymer upcycling strategies that utilize low-temperature and selective catalytic and engineering approaches to obtain high-value and functional materials.
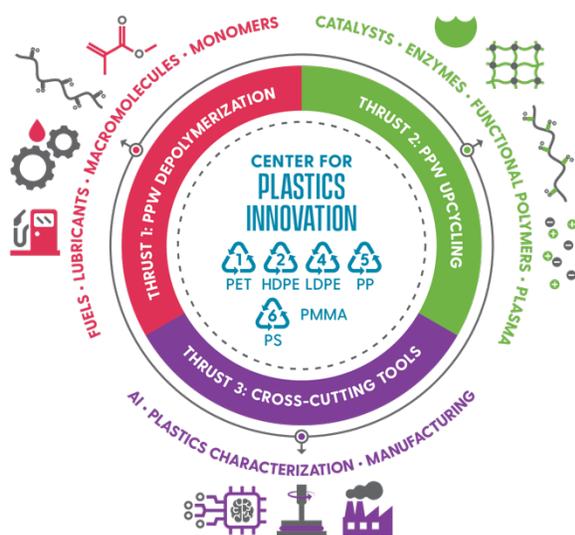
Description: We will leverage our expertise in catalysis (synthetic, biological), macromolecular science and engineering, additive manufacturing, data science and artificial intelligence (AI), systems engineering, and computation to address three distinct thrusts: (1) tunable heterogeneous catalysts and microwave (MW) energy for the conversion of PPW into fuels, lubricants, precision macromolecules, and monomers, (2) functionalization and upcycling using *a)* plasma-assisted, catalysis and surface treatment, *b)* photoredox-catalyzed decarboxylation, and *c)* enzymatic routes to generate high-value monomers and polymers, and (3) new cross-cutting tools driven by AI, macromolecular characterization, and additive manufacturing to enable scientific advances. *CPI* will overcome distinct challenges related to PPW upcycling, including *PPW diversity and heterogeneity*; *optimization of macromolecule/catalyst interactions in melt*; *selectivity control*; and *development of correlations between plastics manufacturing, chemical recycling, and macromolecular physical properties.*

Methods: We will fabricate *hierarchical, multiscale materials* to study interactions between complex PPW and multifunctional catalysts, employ *real-life PPW* to validate our fundamental studies, utilize *bioenzymatic* and *chemical transformations* to add value to polymers waste, design *energy-efficient MW technology* and innovative *plasma-functionalization* strategies, develop *multiscale modeling* and *data mining* tools to enhance predictive capabilities, target *system-level* approaches, harness *new polymer chemistry* and *manufacturing* techniques for valorization, and develop advanced *spectroscopic techniques* for complex media – all to form a basis for significant advances in PPW upcycling.

Impact: We will transform the current high-energy/low-value landscape of polymer recycling towards highly efficient polymer upcycling strategies. Mechanistic insights in MW-assisted, low-temperature catalysis for depolymerization in the melt, plasma-assisted functionalization approaches, photoredox decarboxylation strategies, and enzyme engineering to valorize PPW will provide immense fundamental knowledge. Cross-cutting tools and processes will impart long-lasting impact on science and technology. Furthermore, our systems-level approach will tackle real-life PPW to define new frontiers in research and educational training with direct impact on polymer upcycling, chemistry, manufacturing, catalysis, and data science.

Overarching Goals and Objectives: The complexity of converting PPW presents numerous opportunities for scientific discovery and technological innovation in catalysis, polymers, materials, modeling, and AI science. We will develop a comprehensive program with the **overarching goal** to overcome the fundamental knowledge barriers described above toward advancing PPW chemical recycling and upcycling strategies. *CPI* will focus on PPW spanning from single-stream to multi-component products containing PET, HDPE, LDPE, PP, PS, and PMMA. It will lay the foundation for fundamental science to allow the design of fuels, lubricants, monomers, and macromolecules, as well as enable upcycling with targeted functionality to new polymer structures. The **objectives of *CPI*** include: (1) develop approaches and fundamental tools applicable to the upcycling of real PPW, with a strategic focus on enabling mixed-stream transformations in varied material form factors (*i.e.,* solutions, melts, and surfaces); (2) educate the future U.S. workforce for relevant industries; and (3) enable PPW upcycling innovations *via* technology transfer, licensing, and start-up formation.

Center for Plastics Innovation (CPI)	
University of Delaware	LaShanda Korley (Director), Thomas Epps, III (Deputy Director), Stavros Caratzoulas, Wilfred Chen, Hui Fang, Aditya Kunjapur, Raul Lobo, Michael Mackay, Eleftherios Papoutsakis, Dion Vlachos, Mary Watson
Oak Ridge National Laboratory	Stephanie Galanie, Josh Michener
University of Chicago	Juan De Pablo
University of Massachusetts, Amherst	Peng Bai
University of Pennsylvania	Raymond Gorte

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 C_4-C_6 shale gas derivatives. We have established three 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
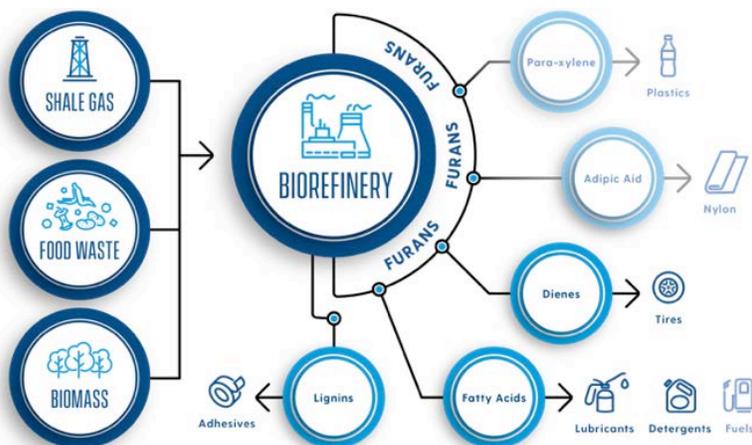
CCEI's research portfolio.

chemistry of furans with (di)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. Furthermore, we attempt feedstock diversification and utilization of renewable or inexpensive carbon sources, such as lignocellulose, food waste, fatty acids and/or shale gas.

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 are extending the use of *P-zeosils*, a new class of catalysts discovered by CCEI that consist of phosphoric acid supported on inert, all-silica zeolites. P-zeosils are Brønsted acids distinct from typical aluminosilicate zeolites with dynamically evolving P-speciation. 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 metal organic frameworks.

The synthesis of bio-renewable oleo-furan-sulfonate surfactants with unique properties requires 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 questions regarding their distribution in porous materials, reaction mechanisms and active site catalyst design in order to accelerate the dehydration of acylating acids and prevent through water management side reactions on strongly acidic sites. Pore design for the control of

the distribution, orientation and diffusion of bulky amphiphiles within porous materials adds one more layer of complexity that we are addressing.

The design and synthesis of macromolecules require the synthesis of multi-functional catalysts for the conversion of furfural derivatives to alkylfurans via hydrogenolysis (surfactants) and for the decyclization-hydrodeoxygenation of the backbone furans to poly-alpha-olefin lubricants. To that end, we are pursuing fundamental research in numerous directions: (1) We are leveraging inverse metal/metal-oxide catalysts as they can possess various sites that cooperate in complex ways. (2) We are exploring selective C-O bond activation and hydrodeoxygenation (HDO) catalysis. Success lies in balancing the C-O bond scission and vacancy formation rates in the presence of H_2. (3) We are exploring use of olefins and dienes from shale gas to produce functionalized substrates, such as aldehydes and acids, for hydroxyalkylation/alkylation chemistry and acylation chemistries.

Catalysis Center for Energy Innovation (CCEI)	
University of Delaware	Dionisios Vlachos (Director), Eric Bloch, Stavros Caratzoulas, Marianthi Ierapetritou, Yan Jin, Raul Lobo, Marat Orazov, Donald Watson, Weiqing Zheng
Stony brook University / Brookhaven National Laboratory	Anibal Boscoboinik, Anatoly Frenkel
California Institute of Technology	Mark Davis
Carnegie Mellon University	Lynn Walker
Columbia University	Jingguang Chen
John Hopkins University	Michael Tsapatsis
University of California Santa Barbara	Song-I Han, Susannah Scott, Phillip Christopher
University of Connecticut	Ioulia Valla
University of Maryland	Dongxia Liu
University of Massachusetts Amherst	Wei Fan, Omar Abdelrahman
University of Minnesota	Paul Dauenhauer
University of Pennsylvania	Raymond Gorte, Christopher Murray

Contact: Dionisios G. Vlachos, CCEI Director, vlachos@udel.edu
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Center for Molecular Magnetic Quantum Materials (M^{2}QM)
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^{2}QM.

With an overarching goal of *turning molecular magnets into quantum materials useful for both quantum computing and quantum current conventional devices,* M^{2}QM 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^{2}QM 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 (^2H, ^{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, Xiao-Xiao Zhang
Florida State University	Stephen Hill (Deputy Director), Michael Shatruck
University of Central Florida	Talat Rahman
Los Alamos National Laboratory	Vivien Zapf
California Institute of Technology	Garnet Chan
University of Texas at El Paso	Mark Pederson

Contact: Hai-Ping Cheng, Director, m2qm.efrc@phys.ufl.edu
352-392-6256, www.efrc.ufl.edu

Quantum Sensing and Quantum Materials (QSQM)
EFRC Director: Peter Abbamonte
Lead Institution: University of Illinois, Urbana-Champaign
Class: 2020 – 2024

Mission Statement*: To develop three new quantum sensing techniques—scanning qubit microscopy, two-electron Einstein-Podolsky-Rosen (EPR) spectroscopy, and nonlinear x-ray optics—and use them to study local and nonlocal quantum observables in quantum materials.*

The EFRC on Quantum Sensing and Quantum Materials (QSQM) will apply new types of quantum sensing technique to measure and correlate local and nonlocal quantum observables in three families of quantum materials central to DOE's energy mission: exotic superconductors, topological crystalline insulators, and strange metals.

To carry out this mission, we plan to develop three new, cutting-edge quantum sensing instruments. The first is a **scanning qubit microscope (SQM)**, which consists of a single flux qubit integrated into a tip that may be scanned over the surface of a material, as illustrated in Fig. 1. The state of a qubit is exceedingly sensitive

Figure 1 Conceptual illustration of a scanning qubit microscope, which consists of a tip-mounted flux qubit that may be scanned in close proximity to a heterogeneous quantum material.

to charge and flux noise, making an SQM a highly sensitive probe of local charge and spin fluctuations near surfaces or interfaces.

The second instrument is a **two-electron EPR spectrometer**, which uses correlated Einstein-Podolsky-Rosen (EPR) pairs of electrons to reveal hidden global interactions in materials. This instrument, illustrated in Fig. 2, can be operated in two different modes. In the first, double photoemission, the sample is illuminated with ultrafast XUV photon pulses that eject correlated electron pairs from the surface. The angle-and energy-distribution of these pairs can be used to reconstruct the anomalous Green's function of the material. In the second, two-electron Rutherford scattering, time coincident ultrafast electron bunches are scattered off one another near a material surface, revealing the renormalized interaction between quasiparticles.

The third, **x-ray four-wave mixing** instrument uses correlated pairs of photons to measure the nonlinear optical response of materials at nonzero momentum. Illustrated in Fig. 3, this experiment is carried out at a free-electron laser facility, such as the LCLS
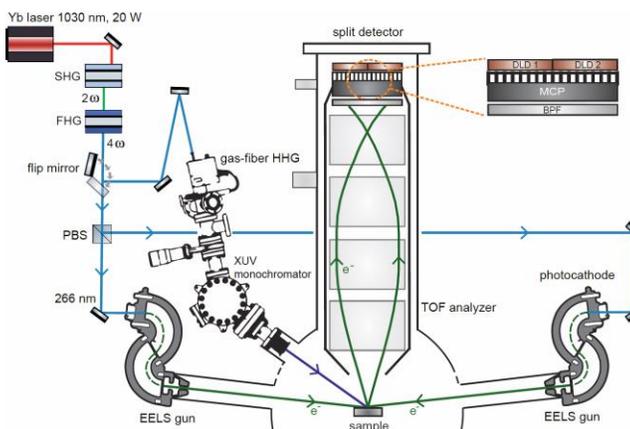
Figure 2 Conceptual illustration of a two-electron, time-of-flight, Einstein-Podolsky-Rosen spectrometer, which uses coherent pairs of electrons to detect valence band phenomena in quantum materials.

facility at SLAC National Accelerator Laboratory. This instrument will directly measure the global geometry of Hilbert space of a material, revealing previously undetected topological phenomena, and will also identify new broken symmetries in materials that reveal exotic and previously unobserved phases of matter.

The new quantum sensing techniques we develop will enable us to unravel the most compelling mysteries of quantum materials. Scanning qubit microscopy will allow us to detect not only localized electronic states but also sources of dissipation and dephasing in materials

Figure 3 Conceptual illustration of an x-ray four-wave mixing experiment, which uses coherent pairs of photons to measure the nonlinear optical properties of quantum materials at nonzero momentum.

over a wide frequency range. It will measure the frequencies of excitations and fluctuations in materials with unprecedented sensitivity. It will enable us to determine whether corner states and hinge states exist in higher-order topological insulators, whether new types of zero-energy states emerge at domain walls in unconventional superconductors, and reveal how strange metals screen charge.

Two-electron EPR spectroscopy will improve our scientific understanding of all materials in which interactions play a nontrivial role, including magnetic materials, strange metals, doped Mott insulators, interacting topological phases, and charge and spin density wave materials. It can directly reveal the pairing boson in most unconventional superconductors, as well as the mechanism behind other types of Fermi surface instabilities in quantum materials.

Nonlinear optics is widely known to be a highly sensitive probe of broken symmetries in materials. However, such measurements have always been restricted to small momenta. Our momentum-resolved x-ray nonlinear optics instrument will allow us to detect new categories of phenomena including the predicted nonlinear axion optical response in topological crystalline insulators, and whether charge carriers in strange metals flow independently or via collective hydrodynamic transport.

Together, these schemes will provide new knowledge about interacting and topological materials, facilitate the discovery of new materials, and define new directions for future spectroscopic probes.

Quantum Sensing and Quantum Materials (QSQM)	
University of Illinois, Urbana-Champaign	Peter Abbamonte (Director), Barry Bradlyn, Tai C. Chiang, Taylor Hughes, Angela Kou, Vidya Madhavan, Fahad Mahmood, Nadya Mason, Philip Phillips, Daniel Shoemaker, Dale Van Harlingen
SLAC National Accelerator Laboratory	Tom Devereaux, Benjamin Feldman, Kathryn Moler, David Reis, Zhi-Xun Shen, Mariano Trigo
University of Illinois, Chicago	Dirk Morr

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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 and artificial intelligence 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.

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

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

- Concerted experimental/computational approaches to (i) make and interpret structural and spectro-scopic 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.

Inorganometallic Catalyst Design Center (ICDC)	
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Clemson University	Rachel Getman
MIT	Heather Kulik
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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 – 2024

Mission Statement: *To develop the chemical understanding and hierarchical structure motifs needed to create materials for effectively immobilizing nuclear waste species in persistent architectures.*

The Center for Hierarchical Waste Form Materials (CHWM) is organized as an integrated, multi-disciplinary team to develop the fundamental science from which advanced waste forms for extreme and dynamic conditions can emerge. A simple and practical definition of a hierarchical structure is that it contains a small-scale structural motif within a larger-scale structure or framework. Conceptually, our hierarchical structures consist of porous structures, either repeating (crystallographically ordered) or non-repeating (disordered), whose cavities are occupied by crystalline or non-crystalline fillers. Examples of materials in this work include crystalline salt inclusion materials (SIMs), metal-organic frameworks (MOFs), porous silica (including Prussian-blue and silver salt functionalized versions), and hollandite mineral structures with molecular tunnels. The CHWM will apply the unique understanding and predictive capabilities developed over the previous four years to make the new discoveries and advances needed to design waste forms with superior stability and performance across extremes in time and environment. The synthesis of such hierarchical structures will be achieved by exploiting previously underutilized or unrealized chemistries.

Hierarchical materials can be tailored from the molecular/crystal lattice through higher scales, offering opportunities to immobilize specific radionuclides in an optimized matrix. Accomplishing such goals requires a fundamental understanding sufficient for effective design of hierarchical materials with tailored properties. Such hierarchical materials can conceivably isolate targeted radionuclides, such as the transuranic elements plutonium, neptunium and americium, more effectively by tolerating higher waste atom content together with lower treatment cost and more rapid processing. Through advances in integrated experiment and modeling, the CHWM, during its first four years, made significant discoveries across a spectrum of hierarchical materials that may provide the foundational science from which researchers can design, manipulate, and ultimately control such materials functionality.
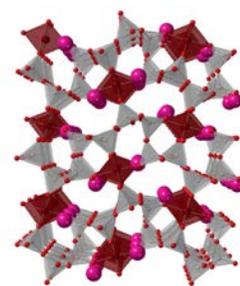
Structure of a new plutonium containing waste form: $Cs_2PuSi_6O_{15}$, Pu (cyan) Si (grey) O (red) Cs (purple).

Novel waste forms are likely to have a transformative impact in reducing the environmental and financial costs of remediation for specific classes of waste. These include the by-products and secondary waste streams that are a result of inefficiencies in current parent processes (e.g., volatile species, poorly retained elements.) and also smaller volume wastes that do not warrant the resources to develop industrially-scalable processes. In addition, there are wastes that are troublesome for known technologies (e.g., those

with low solubilities, limited chemical stability.). Fortunately, recent technological advances in materials synthesis, computational modeling, and advanced characterization now provide tools with which to efficiently pursue innovative waste form development that was previously not possible or prohibitively costly. Hierarchical materials are one of the most promising classes of materials that have the potential to complete the suite of technical approaches that will provide for cost-effective stewardship of the nation's processed nuclear waste.
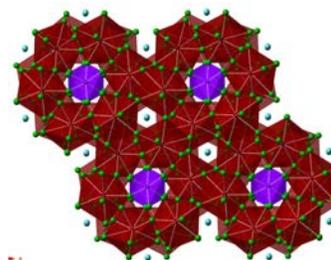
Structure of $Na_3AlPu_6F_{30}$ composed of a $Pu_6F_{30}^{6-}$ framework (red) MF_6 (purple), F^- (green) Na^+ (blue), exhibiting high waste atom content.

The development of new classes of materials to meet current and future Environmental Management challenges that lie beyond the capability of conventional waste forms requires a fundamental understanding of their synthesis, stability, and the transport phenomena occurring within them. Those advances can only be achieved through collaborative efforts among integrated teams such as those present in the CHWM, applying and developing cutting edge materials science. The CHWM is uniquely positioned for this task, benefitting from our established synthetic techniques integrated with the means for structural and compositional modifications, characterization and property measurements, and advanced theory and modeling across multiple length scales. This successful core team, now with the addition of international leaders in radiation effects in materials, will enable advances in the fundamental understanding of novel hierarchical material systems, ultimately leading to structural motifs that can effectively immobilize select nuclear waste species in persistent architectures.

The CHWM will target three research directions to achieve new waste forms: 1. Model, synthesize, and demonstrate the stability of transuranic-containing hierarchical materials; 2. develop direct and indirect synthetic routes for element specific structure motifs; and 3. understand transport in multi-scale porous and hierarchical materials

Center for Hierarchical Waste Form Materials (CHWM)	
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Pacific Northwest Laboratory	Shenyang Hu
Alfred University	Scott Misture
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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.

Integrating Framework

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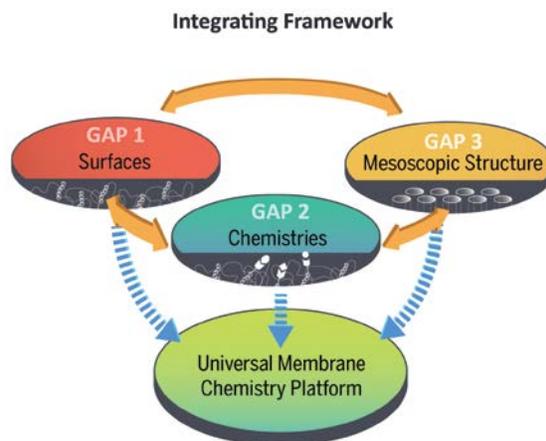
M-WET scientific framework, illustrating the Gap Attack Platforms (GAPs) and Integrating Framework.

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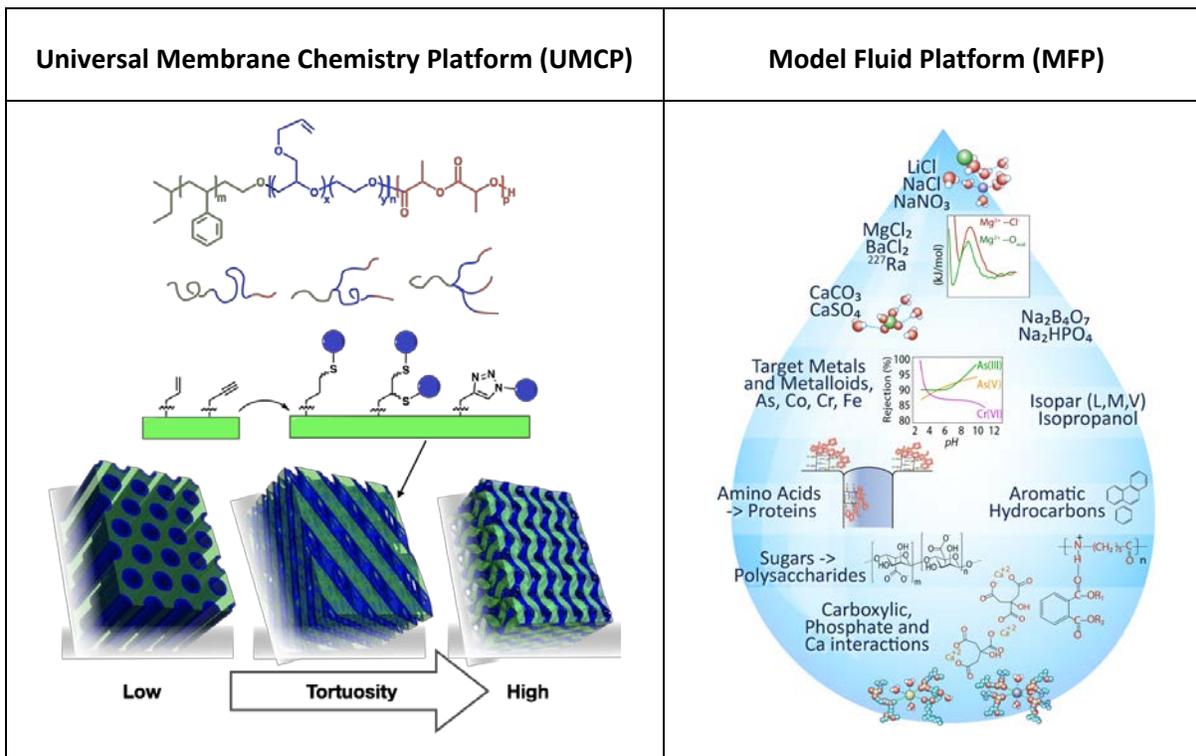
M-WET's Universal Membrane Chemistry Platform and Model Fluid Platform.

Materials for Water and Energy Systems (M-WET)	
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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 geo-inspired materials with repeatable hierarchical heterogeneity and develop an understanding of transport and interfacial properties of fluids confined within these materials.*

Today, energy recovery from the subsurface accounts for more than 80% of the global energy use according to the U. S. Department of Energy, Quadrennial Technology Review. More than 50 billion cubic meters of fresh water are consumed annually for energy production which is unsustainable. With this pressing need for next-generation technologies for a sustainable energy future, the central MUSE research mission of developing a fundamental knowledge of fluid behavior at complex solid interfaces is now as relevant as ever. Interactions of fluids at solid interfaces are key to understanding the thermodynamic, transport, mechanical, and electronic properties of fluids and materials in applications spanning energy storage and production, basic separations, catalysis and carbon capture. There is considerable evidence that the known laws of adsorption, reaction, phase transitions, and flow do not hold for fluids confined in porous materials at the nanometer scale. Thus, new or modified laws must be created based on sound experimental measurements to improve the predictive capability of fundamental models at multiple scales.

MUSE brings together a multi-disciplinary team to address these very challenges by establishing 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. The defining objective of the MUSE EFRC is to address key scientific knowledge gaps on the origins of anomalous flow, thermodynamic, reactivity, and mechanical behaviors of confined fluids in architected materials. Some of the most important challenges include the design of geo-inspired architected materials with precisely defined heterogeneity, detailed insights on the dynamic evolution of siliceous interfaces, the effect of pore and interfacial chemical controls on single and multi-phase flow and nanomechanics, a detailed knowledge of fluid phase behavior in confinement, and bridging measurement and computational scales to probe complex fluid interactions in nanoscale environments.

MUSE is organized into five distinct but highly interrelated research thrusts: (1) *Material Synthesis:* Develop robust geo-inspired architected materials with predictable, hierarchical porosity and surface chemistries; (2) *Properties Measurement:* Measure and understand anomalous thermodynamic, flow, reactivity and mechanical behaviors of confined fluids in hierarchical porous materials; (3) *Dynamic Measurements:* Elucidate the dynamic evolution of chemical structure and pore morphology of solid-fluid interfaces in geo-inspired materials using advanced national core facilities; (4) *Nano-Mechanics:* Probe anomalous deformation, chemo-mechanical coupling and material failure mechanisms due to surface interactions and heterogeneity; and (5) *Modeling and Simulation:* Create validated, atomistically-informed molecular dynamic simulations of materials and fluids at realistic conditions.

Figure 1 shows how these five thrusts are centered around exploring the origins of anomalous thermodynamics, flow, reactivity and mechanical behaviors arising from fluids confined in architected materials through the themes of *interface design*, *thermodynamics in confinement*, *reactivity and mechanics*, and *flow in confinement*. The thrusts and research themes allow rich collaboration between team members, as shown in Figure 2, by the example of ten recent experiments.
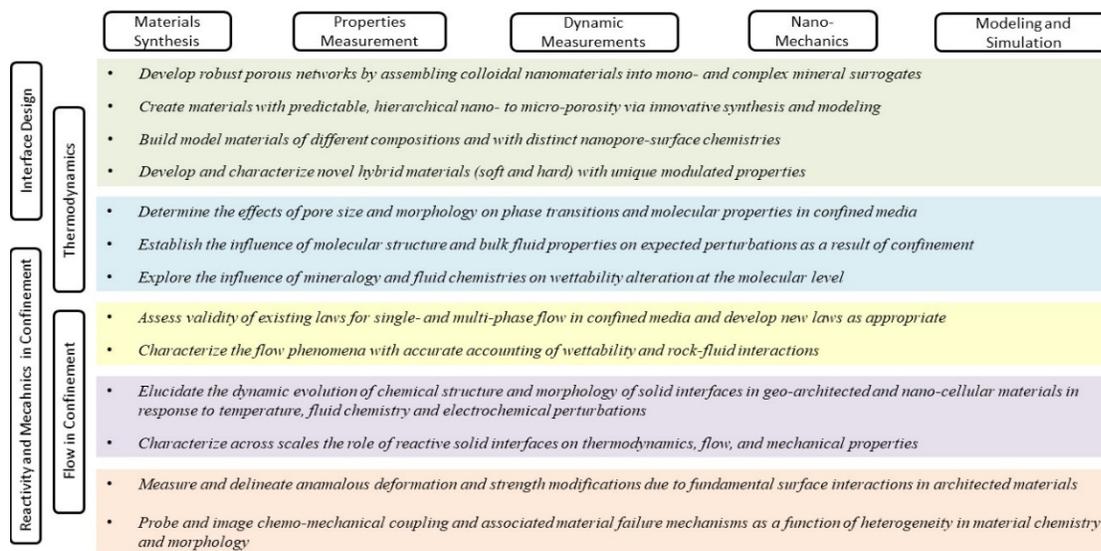
Figure 1. Design, Observations, Modeling and Simulations of Fluids in Architected Geo-Inspired Materials
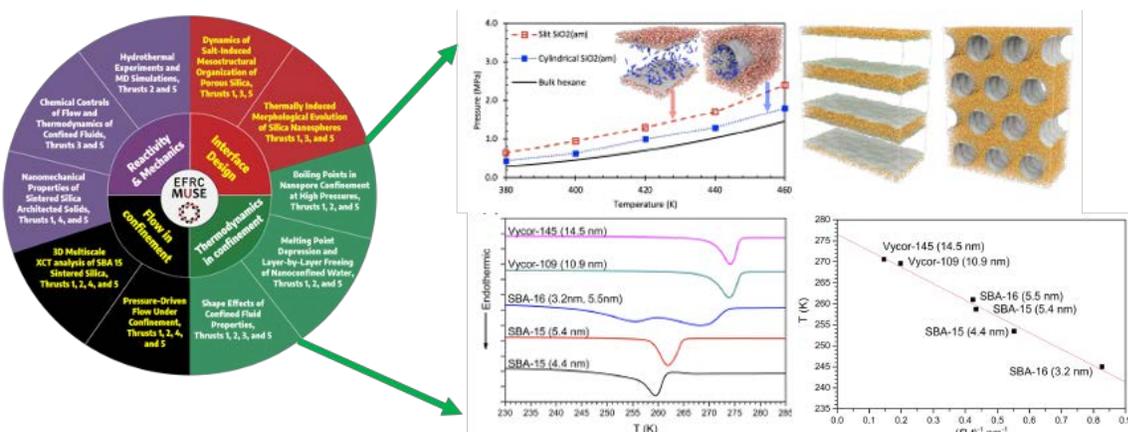
Figure 2. Experiments and Modeling Efforts span different thrusts, allowing rich collaborations.

Multi-scale Fluid-Solid Interactions in Architected and Natural Materials (MUSE)	
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The Pennsylvania State University	Adri van Duin (Thrust 4 Lead)
University of California, Davis	Subhash Risbud
University of Utah	Darryl Butt (Director), Michael Bartl (Deputy Director), Milind Deo (Deputy Director), Michael Hoepfner, Jules Magda, John McLennan (Thrust 5 Lead), 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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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.*
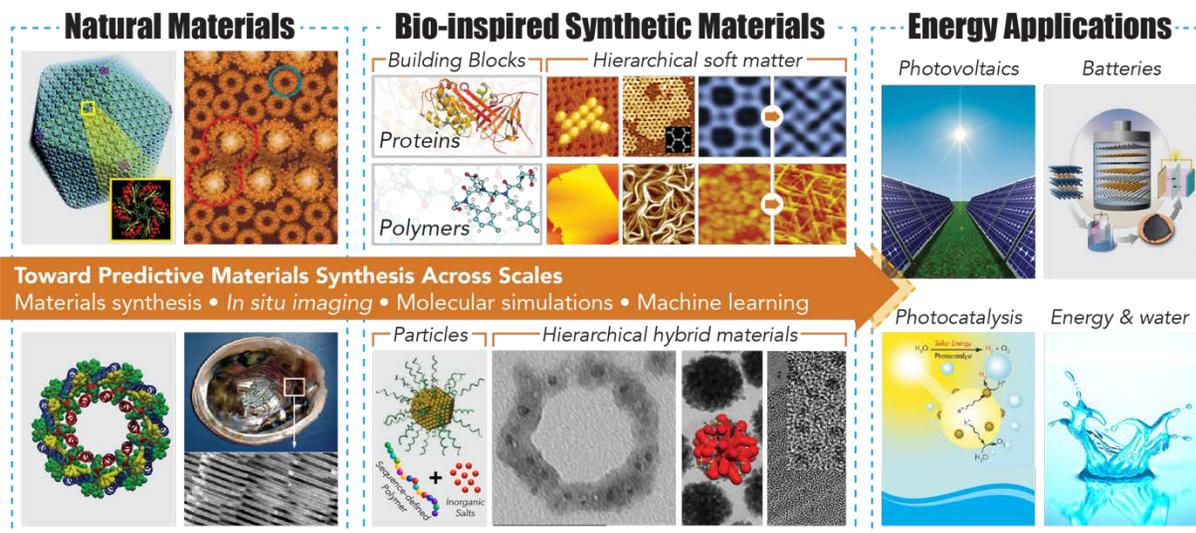
Hierarchical materials offer game-changing opportunities for energy technologies because they enable complex interconnected functions ranging from control of charge and mass transport, to dissipative response to external stimuli and the precise localization of sequential and parallel reactions. Nowhere 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:

Vision: Predictive materials synthesis across all scales
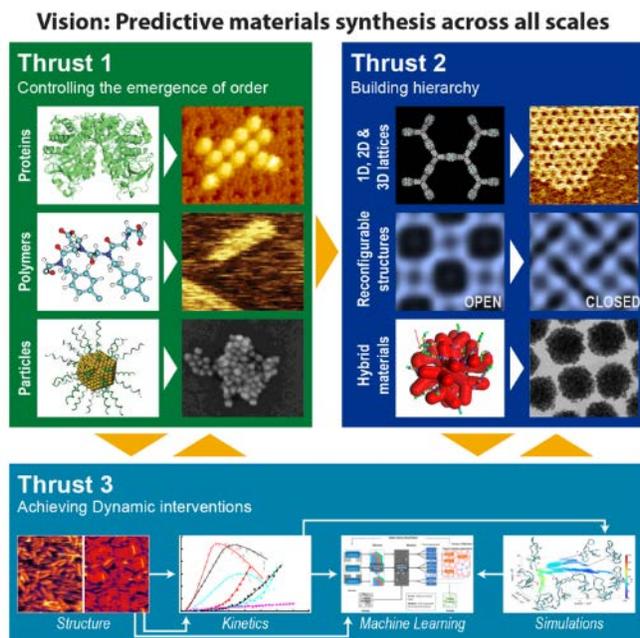
1. To predict how the chemical and/or sequence information of inorganic, polymer and protein building blocks translates into the emergence of order and the outcomes of assembly.

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)	
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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
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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.
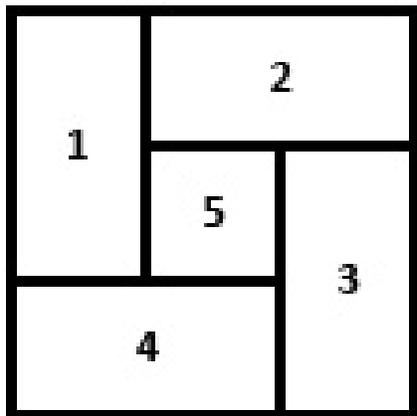
1. Center for the Science of Synthesis Across Scales (CSSAS) EFRC led by François Baneyx at the University of Washington (page 81)

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3. Integrated Mesoscale Architectures For Sustainable Catalysis (IMASC) EFRC led by Cynthia Friend at Harvard University (page 21)

4. Center for Performance and Design of Nuclear Waste Forms and Containers (WastePD) led by Gerald Frankel at the Ohio State University (page 41)

5. Bioinspired Light-Escalated Chemistry (BioLEC) led by Gregory Scholes at Princeton University (page 51)