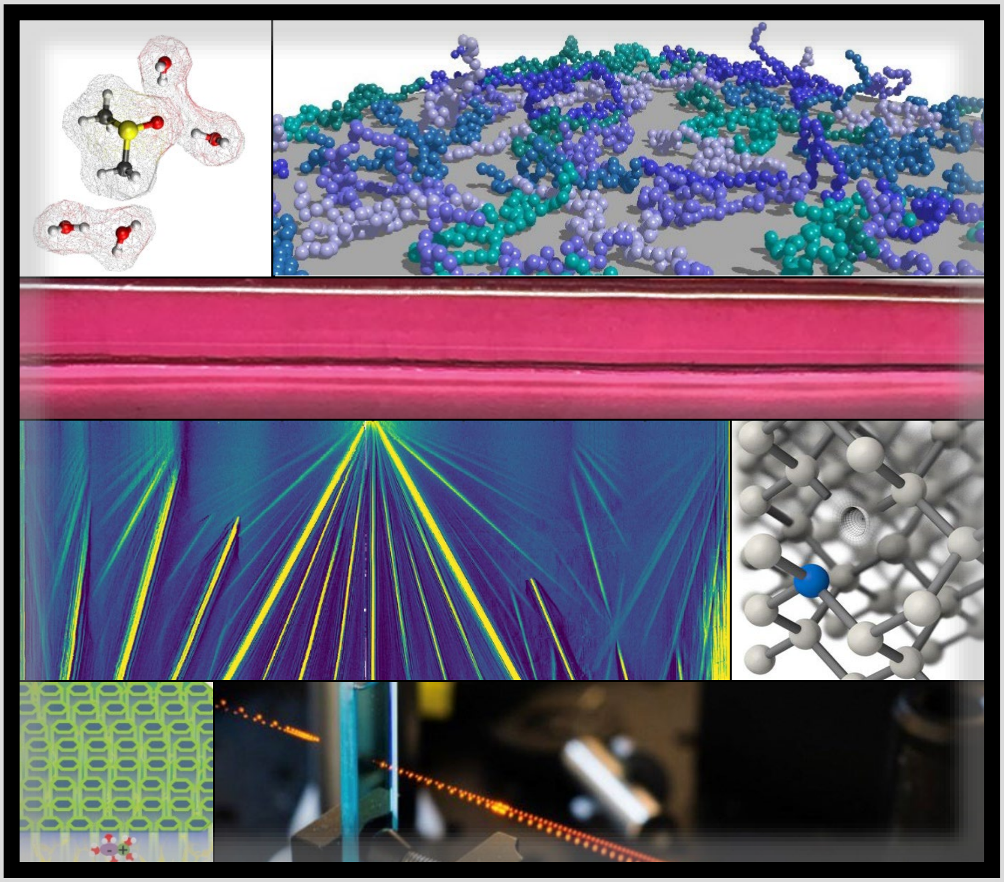
Energy Frontier Research Centers

Technical Summaries



August 2023
https://science.osti.gov/bes/efrc

U.S. DEPARTMENT OF ENERGY | Office of Science

INTRODUCTION

Department of Energy (DOE) Energy Frontier Research Centers (EFRCs) is a basic research program funded by the Office of Basic Energy Sciences (BES) that brings bring together creative, multidisciplinary, and multi-institutional team of scientific researchers to address the toughest grand scientific challenges at the forefront of fundamental energy science research. These centers provide the foundation for collaborative efforts that targets both discovery science and use-inspired basic research, and they aim to address priority research directions and opportunities identified by a series of BES workshop and roundtable reports. These centers take full advantage of powerful new tools for characterizing, understanding, modeling, and manipulating matter from atomic to macroscopic length scales. They also train the next-generation scientific workforce by attracting talented students and postdoctoral researchers interested in energy science.

Since its inception in 2009, there have been 104 centers across 41 states, including the District of Columbia. Biennial funding opportunities for 4-year awards started in 2014. The 2022 funding opportunity marks the 6th class of these Energy Frontier Research Centers awards, with a targeted competition in clean energy sciences, transformative manufacturing, microelectronics, polymer upcycling, carbon dioxide reduction, and quantum information science. The 2022 open competition resulted in 16 new four-year centers, 17 four-year renewal, and 10 two-year extension awards to existing centers. These centers will join the 8 continuing EFRCs awarded in 2020 that focus on environmental management, microelectronics, polymer upcycling, and quantum information science to make 51 active Energy Frontier Research Centers, each one with its own unique mission, to support the Department of Energy's initiatives in advancing basic energy research across the United States.

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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– 2026

***Mission Statement**: To transform how we discover, understand, and harness new topological states of matter.*

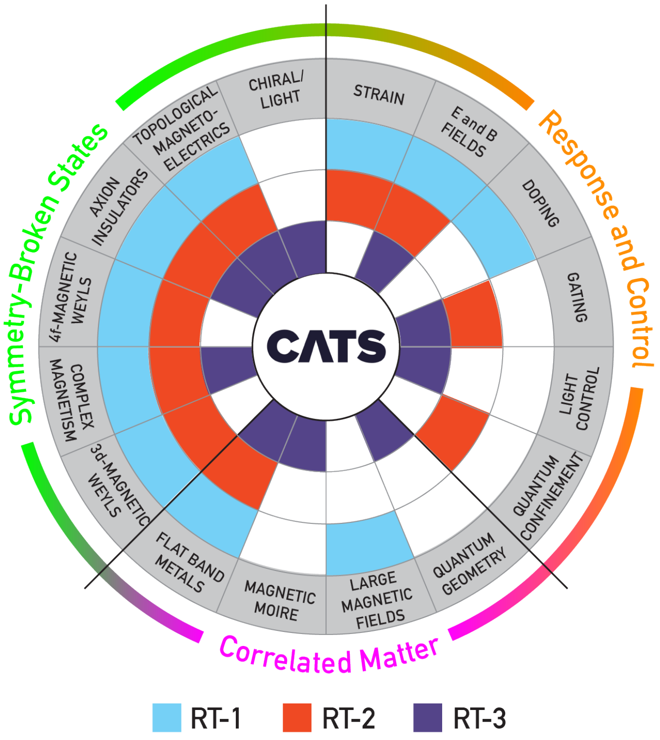
The intersection between topology, magnetism, and electronic correlations is one of the most exciting scientific frontiers that promises to profoundly impact fundamental science and future technologies. Topological materials at this frontier can address major challenges in quantum information sciences and microelectronics by enabling new avenues for controlling charge and spin for low energy switching, by delivering dissipationless currents, and providing new ways to manipulate quantum states. These materials also hold great promise for sensing, detection, and energy harvesting via giant nonlinear transport, optical, and photogalvanic responses. The Center for the Advancement of Topological Semimetals (CATS) will achieve its mission through a highly collaborative fundamental research program, with the vision to provide the innovation that is needed to bring the promises of this exciting frontier to fruition.

CATS groundbreaking successes in the discovery, synthesis, theory, and manipulation of magnetic topological materials and their phenomena inform three, highly integrated research goals for the next four years that both build on and expand the scientific directions within CATS' overall mission:

G1. *Harness the unique properties of symmetry-broken topological materials* – Building on our discoveries of novel phenomena that are caused by the coupling of magnetic order and band topology, we seek to significantly expand this frontier to encompass distinctive spin and charge symmetry-broken ground states in topological materials.

G2. *Discover correlated topological matter* – When combined with topological band structures, strong electronic correlations may give rise to entirely new phases and phenomena. While the discovery of weakly-correlated topological matter has proceeded at an incredible pace, aided by the predictive power of first principles calculations, strongly-correlated topological materials present substantial challenges. CATS' multidisciplinary approach is excellently suited for their theoretical and experimental discovery.

G3. *Control and manipulate the extraordinary responses of topological matter* – Ultimately, the successful adoption of topological matter into future technologies will rely on our



CATS integration. CATS goals are in the outer circle and the principal phenomena and capabilities are colorability to control their phenomena, which, in turn, challenges and advances our scientific understanding. CATS aims to develop unique control parameters available via ultra-high-quality single crystals, thin films, and van der Waals (vdW) assemblies that utilize confinement, twist, pressure, strain, and light to design and control symmetry and topology, aided by the development of theoretical methods to predict novel linear and nonlinear responses based on quantum band geometry.

To attain these goals, CATS is organized into three research thrusts (RTs), centered on three distinct–yet highly interconnected–materials platforms: **(RT-1)** ***Discovery and control of magnetic and correlated topological matter*.** RT-1 emphasizes the prediction and discovery of single-crystals of new magnetic topological materials and understanding their properties and tunability; **(RT-2)** ***Novel topological states in thin films*.** RT-2 recognizes the importance of epitaxial thin films in delivering controllable quantum transport in topological materials; and **(RT-3)** ***Topological magnetism and magnetoelectricity in 2D materials*.** RT-3 focuses on highly tunable vdW assemblies of topological materials with special emphasis on nonlinear optical and transport responses. These thrusts share an overarching ambition to deliver superior materials of exceptional quality, spanning from single crystals to epitaxial thin films to monolayers, respectively.

Integration across CATS is achieved by pursuing a common set of materials, the sharing of theoretical and experimental tools, and the collective pursuit of unifying and cross-cutting scientific concepts linked by broad theoretical insight. Along the way, CATS is developing state-of-the art methods in the growth of new materials, the assembly of functional heterostructures, experimental characterization, and new theoretical concepts and methods. CATS utilizes DOE-supported neutron sources, light sources, nanocenters, and leadership computing user facilities. CATS trains young researchers who will, in the future, make their own discoveries of the fundamental properties of matter.

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Institute for Cooperative Upcycling of Plastics (iCOUP)
EFRC Director: Aaron Sadow
Lead Institution: Ames National 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.

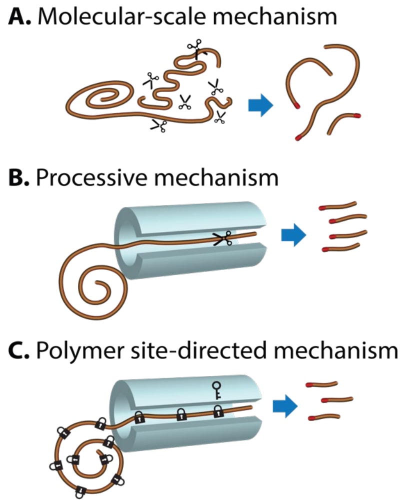

Figure 1. Three mechanistic motifs for polymer upcycling.

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.

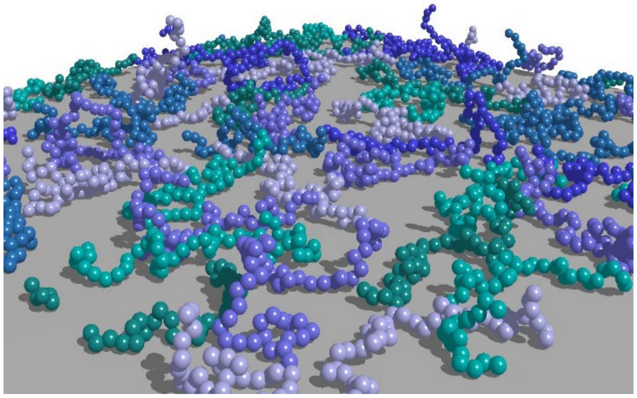

Figure 2. Typical conformation of polymers at a uniformly attractive surface, below the adsorption transition temperature.

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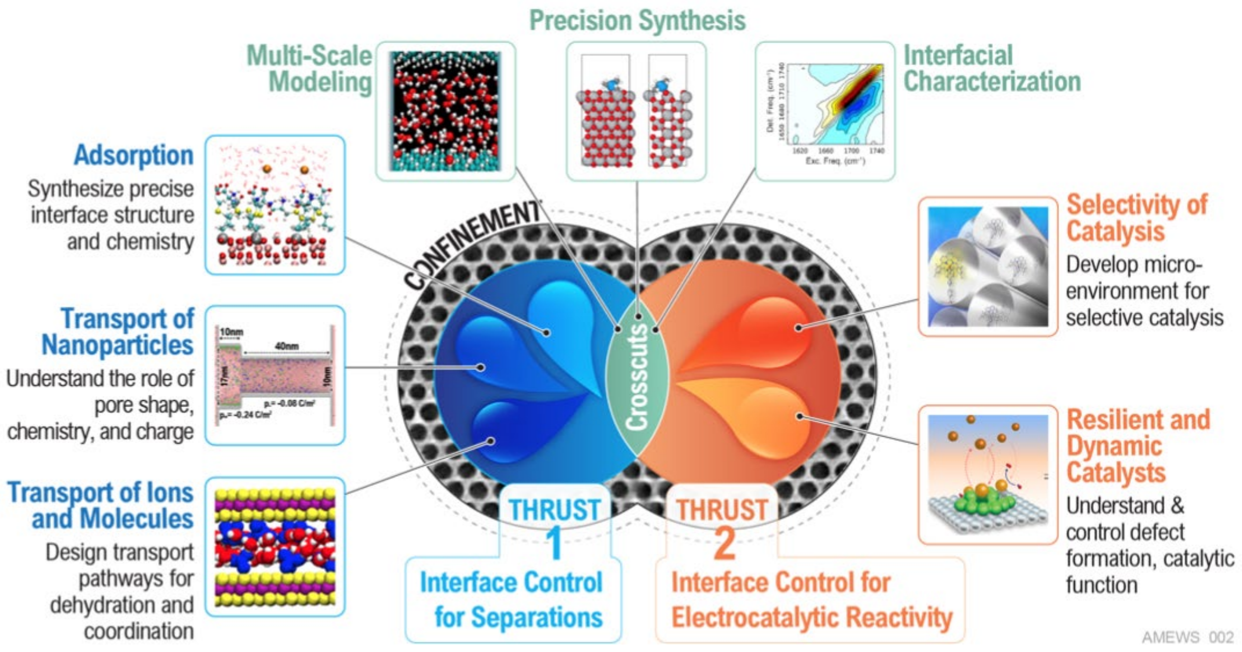
Advanced Materials for Energy-Water Systems (AMEWS)
EFRC Director: Seth Darling
Lead Institution: Argonne National Laboratory
Class: 2018 – 2026

***Mission Statement**: To revolutionize our understanding of aqueous solutes in confined and electrified environments at interfaces.*

Deeper insights into water-solid interfaces are essential for development of innovative and efficient technologies to extract valuable resources from wastewater and to deliver clean water to all people. However, numerous fundamental questions about what happens at water-solid interfaces and why it happens remain unanswered despite decades of study. At the heart of these issues are questions involving the molecular-scale nuances of water's hydrogen bonding at interfaces with electrolyte solutions, interfacial transfer of energy in the form of electrons, and adsorption, transport, and chemical reactivity of solutes at structured, confined, and electrified interfaces. With the emergence of newfound capabilities to experimentally probe and computationally model these complex systems, the chemistry and physics of aqueous solution/solid interfaces is poised for new breakthroughs in understanding.

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

- Design and control transport properties of ions, molecules, and nanoparticles under confinement
- Discover pathways to capture and control release of trace solutes from complex aqueous solutions
- Identify new mechanisms to drive selective electrocatalysis in complex aqueous mixtures
- Predict and synthesize catalysts that are resilient under electro-active aqueous environments



These goals target the two legs of fundamental bases of water-solid chemistry: Interface control for separations and electrocatalytic reactivity.

Water-solid interfaces can result in intriguing properties and activities for both the solution and the solid material in contact with the solution. The organization and dynamics of water, ions, and solutes at the water-solid interface differ significantly from those in bulk solution because of the complex effects of solute-solid interactions, the electrical properties of the interface and solution, and the organization of the water's hydrogen-bond network. It is even more intriguing to describe confining water-solid interfaces that can arise in channels, where there can be two interfaces at distances comparable to the size of a few water molecules. We aim to uncover the principles governing selective adsorption and transport of different target solutes through integrated synthesis, characterization, and theory and modeling.

The research program is further focused on understanding the materials design principles required for effective and selective chemical transformations relevant to water remediation by 1) predicting and synthesizing catalysts that are resilient under electro-active aqueous environments, and 2) identifying new mechanisms to drive selective electrocatalysis in complex aqueous mixtures. To accomplish these scientific goals and address critical knowledge gaps, our team will develop a fundamental atomic- and molecular-level understanding of *dynamic* interfacial structure and concentrate most of our studies on electrocatalysts composed of earth-abundant elements.

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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).

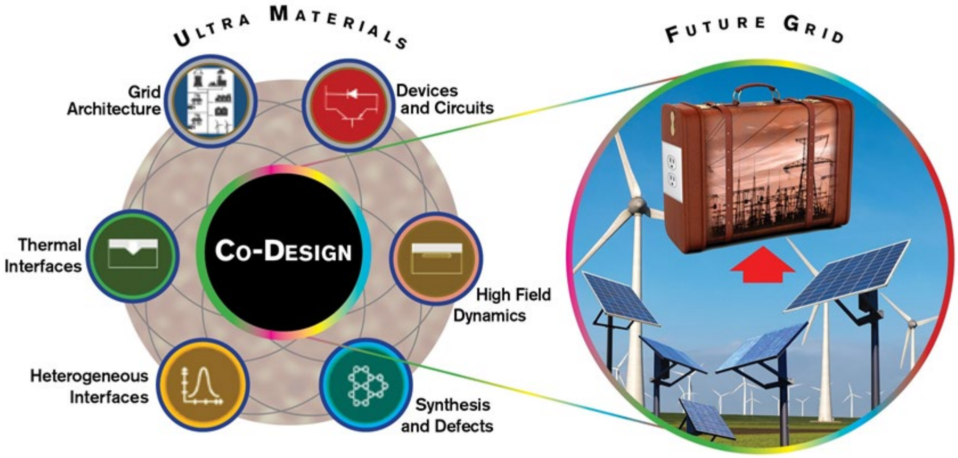

Figure 1. A Future Grid Co-Design Ecosystem will be established to enable communication across all levels of the science and technology.

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.

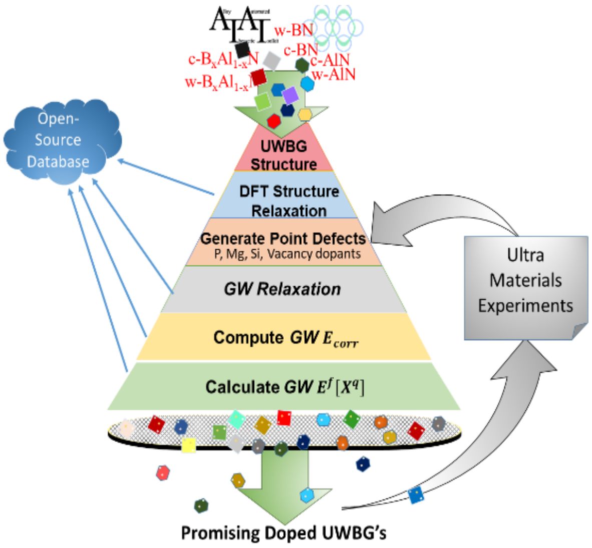

Figure 2. A high-throughput computational approach for identification of low formation energy and shallow donor and acceptor dopants in UWBG's.

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 understood 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.

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Molten Salts in Extreme Environments (MSEE)
EFRC Director: James Wishart
Lead Institution: Brookhaven National Laboratory
Class: 2018 – 2026

***Mission Statement**: To provide fundamental and predictive understanding of molten salt bulk and interfacial chemistry that will establish robust principles to guide the technologies needed to deploy molten salt nuclear reactors.*

Molten salt reactors (MSRs) are a potentially game-changing nuclear reactor technology that is likely to shape the future of nuclear power generation. MSRs could provide a cost-competitive, safe, and more sustainable commercial nuclear power option. MSRs are designed to operate above 500 °C and include molten salts as coolants for solid-fueled reactors, and liquid-salt-fueled reactors containing the nuclear fuel constituents 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.

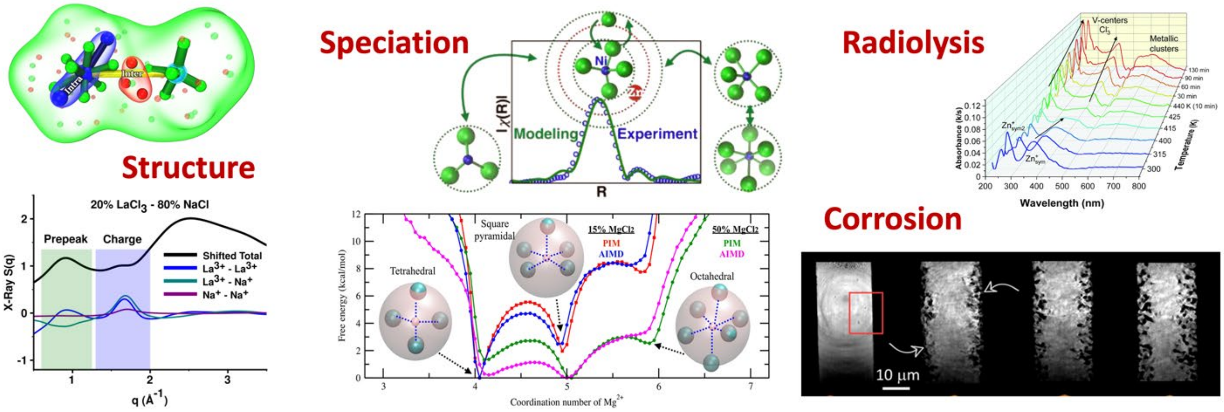
Though made from simple ionic components, molten salts are complex, structured liquids that are subject to intricate physical and chemical processes, particularly in the extreme environments encountered in reactors. Their behavior with solutes, including fuel actinides, and in contact with reactor materials, introduces complex interactions for which fundamental understanding is needed across the range of conditions relevant to reactor operation.

To address this challenge, the integrated, high-level science goals of MSEE over the next four years are:

i. to understand how molten salt atomic-scale structures and interactions control salt dynamical properties and chemical speciation and reactivity, in the bulk and at interfaces, by advancing the state-of-art in experimental and computational methods;
ii. to develop a mechanistic understanding of metal corrosion processes in molten salt systems, ranging from atomistic chemical processes to mesoscale structure and transport; and
iii. to build a comprehensive description of radiation-driven chemical reactivity in bulk fuel salt and at interfaces, to anticipate and mitigate undesirable product formation and material degradation in MSR systems.

MSEE pursues these goals through three synergistic thrusts and one crosscutting theme. The purpose of Thrust 1 (*Molten Salt Structure, Dynamics and Properties*) is to understand the structure and dynamics of neat salts and salt mixtures in the bulk phase across a range of concentrations and temperatures. The purpose of Thrust 2 (*Speciation and Redox Processes*) is to identify and predict the multi-faceted behaviors of the accumulating fission and corrosion products in the salt. The purpose of Thrust 3 (*Interfacial Phenomena*) is to investigate the structure and reactivity of molten salt interfaces with metals and model materials. The purpose of the crosscutting theme of *Radiation-Driven Processes* is to obtain mechanistic understanding of how ionizing radiation changes the speciation of molten salt constituents through redox reactions, leading to new products that alter the properties of the salt system, and driving chemical reactivity at interfaces. These objectives can only be accomplished by closely-coordinated efforts among researchers from multiple disciplines working as a team. Our experience has shown that theoretical insight is essential to properly interpret complex experimental results, for example the coordination environments of technologically important metal ions such as U^{3+} and Ni^{2+}, and counterintuitive changes in the nanoscale ordering of monovalent-divalent salt mixtures with increasing temperature.

A deeper knowledge of 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 greater understanding of the speciation, solubilities and chemical reactivities of dissolved species such as actinides and fission and corrosion products will contribute to better predictions of precipitation and particle formation, participation in corrosion reactions, and ultimately enable stable reactor operation over a wide range of composition, temperature and radiation flux. Improved molecular knowledge of the interfacial processes that drive corrosion, including mass and charge transfer, chemical reactions, and microstructural evolution will suggest ways to mitigate challenges to the endurance of nuclear reactor materials. MSEE will focus on filling these 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 and Systems (BEES2)

EFRC Director: Robert Savinell
Lead Institution: Case Western Reserve University
Class: 2018 – 2026

Mission Statement*: To uncover the transport mechanisms of ions, protons, redox species, and electrons in nano to meso scale structured electrolytes in the bulk and at the electrode-electrolyte interfaces to achieve high energy and power density in next generation energy storage systems.*

Next generation energy storage and chemical transformation technologies require major advances in electrolytes discovery to achieve safer and more efficient performance. Such advances, or breakthroughs, can be in properties or mechanisms not realized in conventional electrolytes. The BEES (Breakthrough Electrolytes for Energy Storage) EFRC, in its first four years (2018 to 2022), studied novel benign electrolytes that are structured at the molecular to meso-scale level. A fundamental understanding of the physical, transport, and electrochemical properties of representative systems in relation to their bulk and interfacial structures was achieved through collaborative experimental and theoretical studies within BEES. In the EFRC BEES2 (Breakthrough Electrolytes for Energy Storage Systems) the overarching goal is to further define design principles of structured electrolytes for achieving breakthroughs in energy density and transport rates of redox species and ions for large scale energy storage devices such as redox flow batteries which are critical to store energy produced from carbon neutral sources such as sunlight, wind, and other renewables.

BEES2 employs a strategy that leverages electrolyte structure to (*i*) conduct protons for proton coupled electron transfer reactions; (*ii*) enhance species transport in the bulk and at interfaces; (*iii*) decouple energy density from conductivity; and (*iv*) control self-assembly in porous electrodes and membranes by expanded efforts in synthesis, computations, and high through-put screening. Two classes of electrolytes, namely CoHBEs (Concentrated Hydrogen-Bonded Electrolytes) and microEmulsions (μEs) are pursued as represented in **Figure 1**. CoHBEs is a coined term that encompasses electrolytes that are structured at the molecular level including deep eutectic solvents and ionic liquids. CoHBEs have high concentration of salts and/or the redox active species, low volatility, and molecular heterogeneity owing to a hydrogen bonding network. On the other hand, μEs are multiphasic electrolytes where liquid droplets containing the redox species are dispersed in a

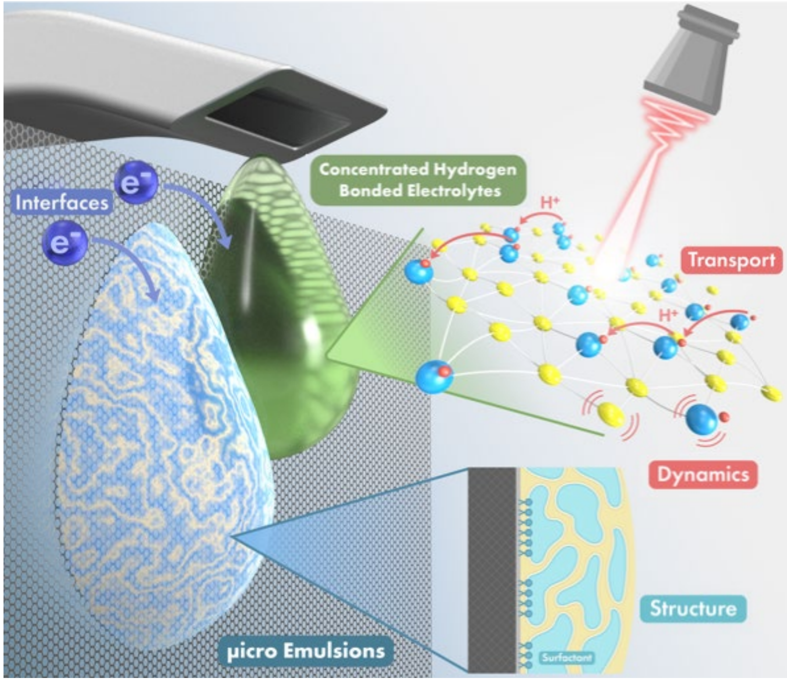

FIGURE 1. Electrolytes of interest to BEES2 EFRC are represented along with bulk (H-bonding network dynamics, proton conduction, structure, *etc.*) and interfacial (electron transfer, redox reactions, *etc.*) properties of importance.

carrier phase to form a single phase; thus, emulsions can decouple electron and charge transfer rates. Further, the chemical and transport properties of µEs can be dynamically controlled by interfacial additives such as nano-scale structured surfactants.

BEES2 is organized into two thrusts: (1) Solvation, Dynamics & Transport and (2) Interfacial Structure & Electron Transfer. In Thrust 1, the main aim is to understand solvation and the coupling of the structural effects of moving ions/protons in the bulk with the dynamics of the transport. Developing these relations requires a hierarchy of experimental and computational approaches that have been established by the BEES team and this established research framework will accelerate discovery of breakthrough electrolytes in BEES2 that are concentrated yet sufficiently conductive. In Thrust 2, the main aim is to understand the voltage induced processes at the electrode-electrolyte interface that include electrolyte interfacial structuring, electrochemical stability, electron transfer mechanism, and kinetics. The understanding of the interfacial redox reactions cannot be complete without an established link to the solvation structure, dynamics, and transport. Therefore, a defined synergistic aim in BEES2 leverages the understanding gained in both thrusts about CoHBEs and µEs to answer the following scientific questions: (1) Can fast proton-conduction networks in CoHBEs be designed and coupled with proton-transfer redox electrode reactions?; (2) For µEs with acidic aqueous phases, how is the underlying proton mobility affected by the presence of maximized oil content?

The fundamental studies proposed by BEES2 will lead to the development of new electrolytes that will overcome the limitations of conventional aqueous and non-aqueous electrolyte systems. BEES2 also will lead to broader scientific and technological impacts beyond flow battery energy storage. It will lead to fundamental knowledge on structure, solvation, transport, and interfacial properties of concentrated hydrogen bonded electrolytes as well as multiphase electrolytes for high efficiency devices for electrolysis, electrochemical materials synthesis, separation processes, sensors, and other applications. The new approaches and models that were developed in BEES and advanced in BEES2 will accelerate the discovery of new electrolyte and more importantly, the breakthroughs in energy density, and transport mechanism and rates.

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Artificially Intelligent Manufacturing Paradigm for Composites (AIM for Composites)
EFRC Director: Srikanth Pilla
Lead Institution: Clemson University
Class: 2022 – 2026

Mission Statement*: to build an AI-enabled inverse design approach for fundamental understanding and integrated material-manufacturing design of advanced polymer composites for improved performance and energy-efficient manufacturing, thereby enabling a smaller carbon footprint, lower structural weight, and lower cost.*

Despite the vast design space of composites, there are significant gaps between the performance, economic and environmental targets and current design and manufacturing approaches. Most egregious are the expensive, long development cycles and the sub-optimal design that waste resources and may adversely affect the environment and climate change. The fundamental cause of such gaps is the lack of detailed understanding of the influence of the material architecture, process methods, and parameters on material microstructure evolution and subsequently the end product's physical, economic, and environmental performance1, which we refer to as the material-process-microstructure-performance (MP2) relationships. The current experimental or analytical material screening approach relies heavily on known material architectures and is a trial-and-error process which largely hinders the material design exploration and optimization capabilities. Such gaps motivate the discovery and construction of a physics-informed, AI-based, inverse design platform that centers on multiscale physics-based models that can capture and predict the parameter space of specific manufacturing processes and material characteristics during fabrication. We envision such a platform that will enable both the discovery of new composites materials forms and relevant new manufacturing methodologies.

The scientific goals of this cooperative research effort are: (1) to unravel the fundamental underpinnings of the MP2 relationship via constructing an uncertainty-aware multi-objective "Digital Life Cycle" (DLC) that represents a suite of seamlessly linked, experimentally converged, high-fidelity models embracing all stages of a composite component's life cycle, linking perceived risk from energy consumption to carbon footprint; (2) to leverage physics-informed AI models and build microservice-based cloud tools to enable inverse composites material architecture and manufacturing process design and in situ diagnosis and control; and (3) to inform and validate the DLC and AI models and implement new material and process designs by exploiting innovative material engineering, characterization, and testing methods. The scientific goals will be achieved via three research thrusts as shown in the three circles in Fig. 1. The green circle is the DLC representing a suite of seamlessly linked, high-fidelity multiscale models for simulating all stages of a polymer composite's life cycle, which also integrates uncertainty quantification and energy, environment, economy (E^3) impact evaluation. The DLC will enable the generation of a large quantity of high-fidelity data for the training of AI models. Equipped with the DLC generated data, in the AI modeling and inverse design research thrust

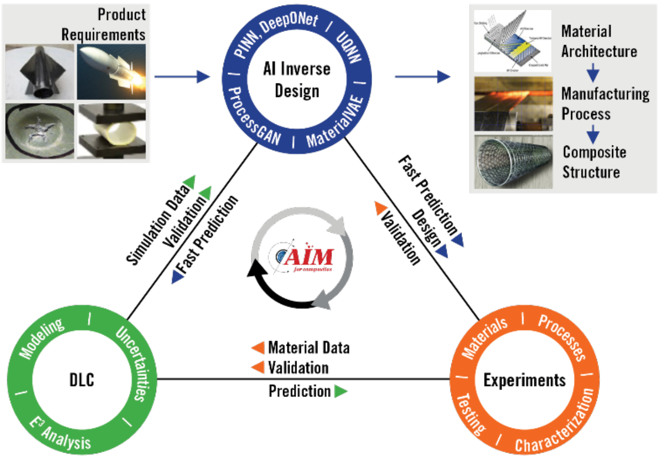

Figure 1. *Project overview: iterative loop among the DLC, AI modeling and Experiments research thrusts, then AI models enable inverse design workflow.*

(blue circle), we will develop new AI models, including physical informed neural networks (PINN) and multiscale deep neural operators (DeepONet), to efficiently map the composite materials' architecture and the manufacturing process to composite components' performance. Next, we will develop a conditional VAE neural network (MaterialVAE) for material inverse design and a conditional VAEGAN neural network (ProcessGAN) for manufacturing process design. Third, we will develop an uncertainty quantification neural network (UQNN) for in situ manufacturing diagnosis and control. By utilizing the experimental facility and capabilities at the Clemson Composites Center, the Center for Manufacturing Innovation at the University of Florida, the Pacific Northwest National Laboratory (PNNL) and other relevant BES facilities and infrastructure, we will conduct material characterization and testing of mechanical, physical, rheological, and morphological properties at nano-, micro-, and macroscales to inform and validate both DLC and AI models and simulations (orange circle of Fig. 1). We will also implement new/hybrid processes that combine existing or new scalable processing routes to create tailored composite micro and macro structures. Finally, the inverse design is performed by the generative AI models. For given performance requirements, the material inverse design is first carried out using MaterialVAE to achieve the target material properties. For each material design candidate, and with the quality and E^3 impact requirements, the manufacturing process inverse design is performed using ProcessGAN. Thus, the optimal composite material and its manufacturing process is obtained as a holistic solution.

Through the proposed research, the AIM for Composites EFRC aims to address the following challenges:

1. The models revealing the MP2 relationship need to be able to capture the material behavior at multiple length scales (impurities, complex compositions), the effects of manufacturing process (phase changes, non-equilibrium characteristics, E^3 impact), and both aleatory and epistemic uncertainties.
2. The possible material architectures and process conditions lead to a vast material and process design space with unknown boundary and few data points, so it is challenging to determine the data sampling strategy and the volume of data to be generated for training ML models.
3. Integration of experimental characterization and testing with model development and validation.
4. How the physical principles will be preserved in ML models for them to represent nonlinear and transient functional properties.
5. Efficient and accurate models that enable in-situ diagnosis and in-process decision making.
6. E^3 impact together with material property, manufacturing quality, and structural performance makes the inverse design multiscale, multi-objective and multidisciplinary.
7. Data fusion and flow among the DLC and ML models, experiments, and the inverse design steps.

Artificially Intelligent Manufacturing Paradigm for Composites (AIM for Composites)	
Clemson University	Srikanth Pilla (Director), Gang Li (Associate Director), Zhen Li, Qiong Zhang, Mik Carbajales-Dale, Feng Luo, James Sternberg
Brown University	George Karniadakis (RT-2 Leader)
University of Florida	Young Huang (ECA Mentor)
Ohio State University	Farhang Pourboghrat (RT-1 Leader), Taejoon Park
South Carolina State University	Nikunja Swain, Biswajit Biswal, Jagruti Sahoo
Savanna River National Laboratory	Dale Hitchcock
Pacific Northwest National Laboratory	Kevin Simmons (RT-3 Leader), Yao Qiao

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Programmable Quantum Materials (Pro-QM)
EFRC Director: D.N. Basov
Lead Institution: Columbia University
Class: 2018 – 2026

Mission Statement*: To discover, characterize and deploy new forms of quantum matter controllable by light, gating, magnetic proximity electromagnetic environment, and nano-mechanical manipulation, effectively programming their quantum properties.*

Discovering quantum phases of matter and controlling their properties is an essential goal of the physical sciences. Quantum phases with controllable properties are required for new electronic, photonic, and clean energy applications needed to address mounting societal demands for fast and energy efficient sensing and information processing and transmission *Quantum materials (QMs*) are appealing platforms for engineering "on-demand" quantum phases because they host interacting many-body electronic states born of the interplay between topology, reduced dimensionality, and correlations. The emergent "quantum matter" exhibits readily tunable quantum effects over a vast range of length, time and energy scales.

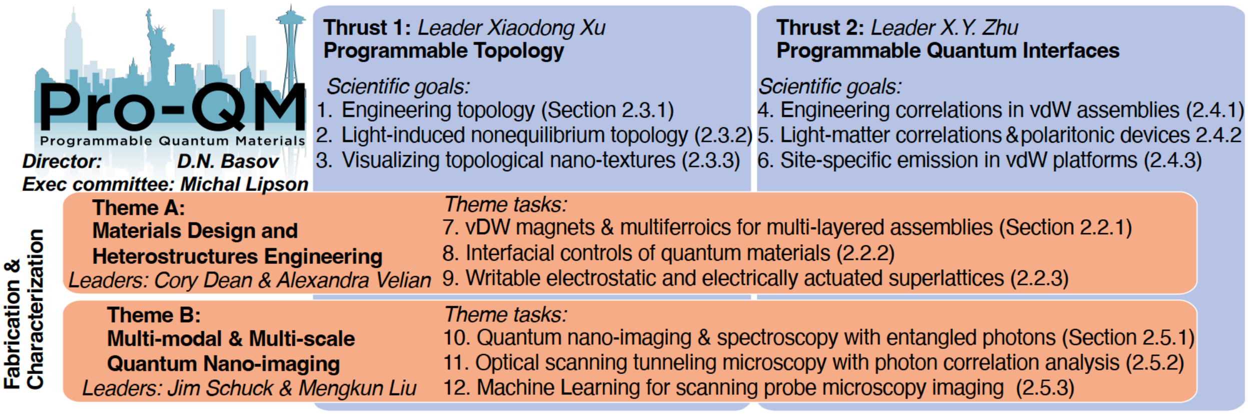
The Pro-QM team is assembled of leading researchers with complementary skill sets who have pioneered some of the key advances in QMs. The effort will be led by Columbia University, in partnership with the University of Washington in Seattle, Brookhaven National Laboratory and the Flatiron Institute in New York City as unfunded partner. The Pro-QM team is comprised of six female physicists, chemists and engineers with *Lipson* and *Velian* serving in leadership positions (Fig.1.1). One third of our co-PIs currently hold tenure track positions.

The scientific goals of the four-year program of Pro-QM are organized in two interdependent Research Thrusts (Fig. 1.1). Thrust 1 will create and image interacting topological states in van der Waals (vdW) quantum materials with emphasis on dynamic manipulation of band topology and on-demand topological spin textures. Thrust 2 will utilize interface phenomena for engineering correlated phases with new forms of electronic/magnetic order and will also create interacting light-matter interfaces in vdW structures hosting quantum effects down to the single-photon level required for robust and scalable quantum hardware. The two thrusts harness and rely on two cross-cutting research themes: Theme A, engineered materials, assemblies and architectures, and Theme B, transformative quantum nano-imaging. The concerted EFRC effort is therefore imperative to make the desired leaps in progress.

Programmable QMs are essential for realizing the revolutionary promise of quantum technology for disruptive advances in information transfer, processing, and sensing, because they enable currently inaccessible properties opening the door to new and, currently unimagined functions.

The Pro-QM team will investigate two-dimensional (2D) van der Waals QMs in view of their outstanding diversity of ultra-manipulatable properties. 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.



Pro-QM
Programmable Quantum Materials
Director: *D.N. Basov*
Exec committee: Michal Lipson

Thrust 1: *Leader Xiaodong Xu*
Programmable Topology
Scientific goals:
1. Engineering topology (Section 2.3.1)
2. Light-induced nonequilibrium topology (2.3.2)
3. Visualizing topological nano-textures (2.3.3)

Thrust 2: *Leader X.Y. Zhu*
Programmable Quantum Interfaces
Scientific goals:
4. Engineering correlations in vdW assemblies (2.4.1)
5. Light-matter correlations & polaritonic devices 2.4.2
6. Site-specific emission in vdW platforms (2.4.3)

Fabrication & Characterization

Theme A:
Materials Design and Heterostructures Engineering
Leaders: Cory Dean & Alexandra Velian
Theme tasks:
7. vDW magnets & multiferroics for multi-layered assemblies (Section 2.2.1)
8. Interfacial controls of quantum materials (2.2.2)
9. Writable electrostatic and electrically actuated superlattices (2.2.3)

Theme B:
Multi-modal & Multi-scale Quantum Nano-imaging
Leaders: Jim Schuck & Mengkun Liu
Theme tasks:
10. Quantum nano-imaging & spectroscopy with entangled photons (Section 2.5.1)
11. Optical scanning tunneling microscopy with photon correlation analysis (2.5.2)
12. Machine Learning for scanning probe microscopy imaging (2.5.3)

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), Cory R. Dean, Milan Delor, James Hone, Michal Lipson, James McIver, Andrew J. Millis, Abhay Pasupathy, Raquel Queiroz, Xavier Roy, P. James Shuck, and Xiaoyang Zhu
University of Washington	Jiun-Haw Chu, David H. Cobden, Alexandra Velian, Di Xiao, and Xiaodong Xu, and Matt Yankowitz
Brookhaven National Laboratory	Valentina Bisogni, and Mengkun Liu

Contact: D.N.Basov, Pro-QM EFRC Director, Columbia University, 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 – 2026

***Mission Statement**: **CABES** seeks to advance the scientific understanding of the fundamental factors governing electrocatalysis and electrochemical energy conversion in alkaline media.*

CABES, the *Center for Alkaline Based Energy Solutions* aims to achieve a detailed understanding of the nature, structure, and dynamics of electrocatalysis in alkaline media and their impact on fuel cell and electrolyzer technologies. CABES integrates theory and computational methods, synthesis of electrocatalysts and novel membrane materials and the development of experimental tools that will provide *in situ*, spatiotemporal characterization of systems under operation. Our programmatic focus and vision are based on 3 fundamental science drivers (SDs):

1. SD-1. What factors govern electrocatalysis in alkaline media?
2. SD-2. How do we understand and control transport in alkaline media?
3. SD-3. What makes energy materials durable in alkaline media?

The center integrates theory and computational methods of catalysis and interfacial structure and dynamics, the synthesis of model (electro)catalytic systems with atomic level control, ionically conducting/transporting ionomers/membranes in contact with electrodes, computational materials science to guide the synthesis of other materials, and the development of experimental tools that will provide *operando*/*in situ*, spatiotemporal characterization of systems under operation. These studies are aimed at greatly accelerating the development of electrocatalysis in alkaline media by generating the fundamental knowledge for the rational development of new materials and architectures. We foresee the center as providing the scientific basis for ushering in an alkaline-based energy system that utilizes abundant elements and is scalable to the needs of society.

CABES addresses fundamental issues of critical needs in energy conversion technologies, guided by the challenges articulated in the relevant DOE reports, Basic Research Needs and Round Tables. CABES SD-based approach revolves around synergistic loops. Fig. 1 presents the loop for SD-1 that integrates materials synthesis and characterization, electrochemical testing, and modeling/data science. Each component within the loop informs the others; i.e., theory may suggest new catalysts to synthesize, and characterization may suggest new structures to simulate and synthesize. With unique characterization tools to examine the structure and strain distributions in catalyst nanoparticles at atomic resolution, and at the nanoscale *in-situ* and *operando*, and electrochemical testing of both components and membrane electrode assemblies, we have information on both structure and properties to guide the synthesis, and test and inform the modeling. This enables us to explore and optimize the synthesis of catalyst nanoparticles in a systematic manner. We also introduce high-entropy materials to increase the catalyst design space and stability options, as the "cocktail effect" from incorporating many different elements leads to unique chemical and electronic environments. The *ab-initio* modelling of surface structure and reaction pathways will greatly benefit from our data science tools for exploring these higher-dimensional spaces.

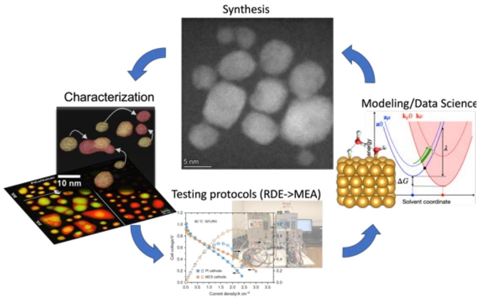

Fig. 1. SD1 synergistic research loop illustrating interactions between/among the different

Science Driver 2 aims to achieve a fundamental understanding of how the architecture, especially of electrolyte/membrane materials, and interfaces within them, influences ion and molecule transport in electrochemical systems under alkaline conditions. The relationships between architecture and transport are investigated through the systematic preparation and study of alkaline exchange membranes (AEMs), correlated ionomers, and the respective interfaces. We have developed novel tools for their study including cryogenic transmission electron microscopy (cryo-TEM) (Fig. 2). We also assess the transport properties that result from these structures.

The aim of Science Driver 3 is to understand what makes energy materials durable in alkaline media. Efforts are organized around fundamental questions that include degradation pathways in alkaline media of membranes and ionomers during transport of OH^- and how can we design and synthesize electrolytes with high hydroxide conductivity yet maintain their long-term durability under operating conditions. We are also focused on understanding interactions of membranes and ionomers with catalysts/supports.

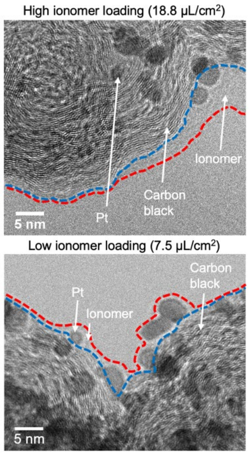

Fig. 2. Non-uniform ionomer distribution revealed by low-dose, cryo-TEM imaging of ionomer-

We have also established a foundation of studies of non-precious metal based electrocatalysts with particular emphasis on metal-nitrogen-carbon, transition metal oxides, perovskites and spinels as well as transition metal nitrides. These studies are providing valuable design criteria for the synthesis of next generation high performance and highly stable electrocatalysts.

CABES has also developed unique experimental capabilities including cryo-TEM for the study of membranes and ionomers in contact with electrocatalysts and supports. We have developed a liquid cell incorporating electrodes on the viewing membrane in a transmission electron microscope.

For studying the role of defects in interface instability we have developed X-ray nano-diffraction and nano-fluorescence analysis to quantify structural defects in catalysts for OER. By leveraging nanoprobe diffraction, we have developed a new tool for localizing, with nanometer resolution, regions with high strain and will extend this method to nanoimaging to examine the impact of strain and defects on electrocatalysis in alkaline media; one of the main research themes of CABES.

We feel confident that CABES will help establish the basis for ushering in and enabling an alkaline-based energy technology society.

Center for Alkaline-Based Energy Solutions (CABES)	
Cornell University	Héctor Abruña (Director), Paul Mutolo (Executive Director), Danielle Hanes (Center Manager), Tomas Arias (SI), Joel Brock (SI), Geoffrey Coates (SI), Robert DiStasio (SI), Brett Fors (SI), Emmanuel Giannelis (SI), Lena Kourkoutis (SI), David Muller (SI), Richard Robinson (SI), Andrej Singer (SI) Jin Suntivich (SI)
Carnegie Mellon University	Kevin Noonan (SI)
Los Alamos National Lab	Piotr Zelenay
National Renewable Energy Laboratory	Bryan Pivovar
Pennsylvania State University	Raymond Schaak (SI)
University of Pennsylvania	Tom Mallouk (SI)
University of Texas, El Paso	Carlos Cabrera (SI)
University of Wisconsin	Emmanouil Mavrikakis (SI)
Yale University	Sharon Hammes-Schiffer (SI)

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Understanding & Controlling Accelerated and Gradual Evolution of Materials for Energy (UNCAGE-ME)

EFRC Director: Ryan P. Lively
Lead Institution: Georgia Institute of Technology
Class: 2014 – 2026

***Mission Statement**: To develop a deep knowledge base in the characterization, prediction, and control of materials evolution in the presence of realistic contaminants, processes, and mixtures to accelerate materials discovery for sustainable production and utilization of H_2 and CO_2*

The overall objective of UNCAGE-ME from 2014-2022 has been to develop fundamental structure-property relations describing how acid gases interact with and induce evolution of adsorbents and catalysts. This was achieved via an interdisciplinary effort that combined novel synthesis, advanced in situ/operando characterization, machine learning techniques and molecular modeling. The creation of structure-property relationships accelerated materials discovery for acid gas separations, conversion, and utilization via these integrated design tools. The research accomplishments of the Center over this 8-year period provided detailed descriptions of the impact of acid gas exposure on metal-oxides, metal-organic frameworks, carbons, supported amines, porous organic cages, and other materials.

In Phase III, we will leverage our Center model and learnings from our acid gas campaigns to accelerate progress in new directions. In line with our Phase III Mission Statement our new focus is on the evolution of materials relevant to clean energy technologies when exposed to realistic contaminants, processes, and complex mixtures. This will be achieved by combining new syntheses, in situ/operando characterization, molecular modeling, and machine learning approaches applied to catalyst, sorbent, and membrane use in clean energy technologies such as H_2 generation and CO_2 capture and conversion.

We will leverage our learnings from Phase I-II to address fundamental aspects of novel materials synthesis, materials stability, and structural evolution that underpin a suite of clean energy technologies relevant to H_2 and CO_2 processing. These include: (i) generation of H_2 via polyelectrolyte membranes (PEMs) and solid oxide electrolysis cells (SOECs); (ii) direct air capture of CO_2; and (iii) electrochemical reduction of CO_2. The name and mission of UNCAGE-ME have been updated to reflect these new focus areas and serve to motivate the Center's research portfolio. These activities are driven by a set of four-year research goals:

1) Elucidate the overarching relationships for process-induced structure and property evolution of functional materials with a focus on separations media and (electro)catalysts.
2) Leverage and advance computational and machine learning techniques to enable fundamental molecular and electronic level predictions of materials interacting with complex mixtures of targeted gases and contaminants.
3) Demonstrate accelerated materials discovery for clean energy technologies via process-materials coupled research.

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 contaminants and non-steady-state conditions under realistic conditions is the norm rather than an exception. In Phase III, we will synthesize novel forms of matter with tailored functional properties and combine in situ/operando molecular spectroscopic studies of both the surface functionalities and bulk structures of materials relevant to the catalytic formation and separations of CO_2 and H_2 under conditions relevant to complex environments. Our experimental findings will be fused with complementary data analytics and multi-scale computational and theoretical modeling of emerging contaminant interactions with functional solids for targeted materials design.

An overview of the various research thrusts and cross-cutting tracks for Phase III is shown in **Figure 1**. Teams comprised of members from each Thrust will address basic scientific questions associated with the technologies described in each track. We have built teams with a broad range of skillsets comprised of senior personnel from each of the three **Research Thrusts** and these teams will be challenged to address fundamental scientific questions related to two **Clean Energy Technology Tracks** (CO_2 Capture & Conversion and H_2 Conversion).

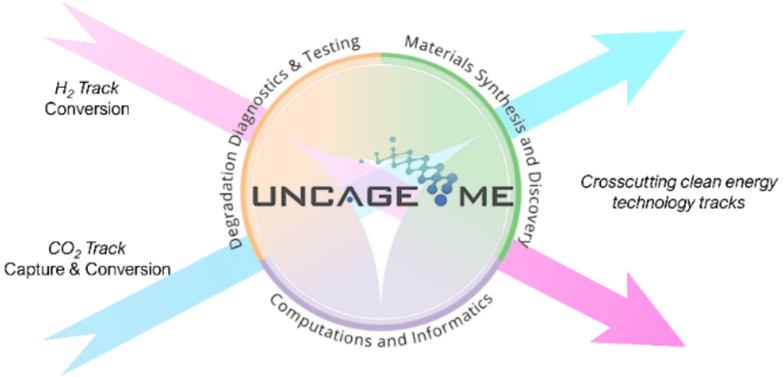

Figure 1: Schematic overview depicting the application of UNCAGE-ME research thrusts (circular emblem) to cross-cutting clean energy technology tracks (arrows). Teams of researchers from each thrust will address specific scientific questions associated with each track.

The Research Thrusts comprise groupings of senior personnel based on expertise. The three thrusts are: (i) Materials Synthesis & Discovery; (ii) Degradation, Diagnostics, & Testing; and (iii) Computation & Informatics. The Materials Thrust focuses on designing, synthesizing, and testing model and new functional materials with tailored properties for clean energy technologies in realistic process streams relevant to H_2 generation, separation, and conversion as well as CO_2 removal from the air and conversion. The Degradation Thrust will focus on advanced in situ/operando structural and performance analysis of model and new materials for H_2/CO_2 processing under relevant conditions. We will leverage existing infrastructure that was purpose-built for UNCAGE-ME Phases I-II as well as some new methods (e.g., Modulation Excitation Spectroscopy (MES), Near Atmospheric Pressure (NAP)-XPS, etc.) for some of the measurements conducted in this thrust. The Informatics Thrust will use advanced computational and data-driven approaches coupled with the experimental molecular-level insights from the other two Thrusts to determine underlying causes of material evolution, predicting this evolution under process-relevant conditions, and ultimately accelerating materials discovery and advancement into technology.

Understanding & Controlling Accelerated and Gradual Evolution of Materials for Energy (UNCAGE-ME)	
Georgia Institute of Technology	Ryan Lively, Krista Walton, Christopher Jones, David Sholl, Sankar Nair, Matthew McDowell, Rampi Ramprasad
Sandia National Laboratories	Tina Nenoff, Jessica Rimsza
Oak Ridge National Laboratory	Juliane Weber, Zili Wu
Lehigh University	Jonas Baltrusaitis, Israel Wachs, Srinivas Rangarajan
University of Alabama	David Dixon
University of Michigan	Eranda Nikolla
University of California Riverside	Kandis Leslie Abdul-Aziz
University of Florida	Coray Colina

Contact: Ryan Lively, Director, ryan.lively@chbe.gatech.edu

Center for Thermal Energy Transport under Irradiation (TETI)
EFRC Director: David Hurley
Lead Institution: Idaho National Laboratory
Class: 2018 – 2026

Mission Statement*: To accurately predict, from first principles, thermal energy transport in actinide materials in extreme environments.*

In nuclear fuel, irradiation-induced defects effectively scatter thermal energy carriers (electrons and phonons), greatly reducing the capacity of the fuel to transport heat to the coolant for eventual electricity generation. For example, in oxide fuels, the thermal conductivity decreases by as much as 70% over the operational lifetime of the fuel. This reduction significantly impacts fuel performance, safety margins, and the amount of usable energy. However, in some special cases, microstructure evolution can lead to local increases in thermal conductivity. For oxide fuels, examples include a reduction in phonon scattering associated with the transformation of faulted loops to perfect loops (a change in the strain field), transformation of a loop ensemble into a line segment (change in defect dimensionality), and defect segregation at interfaces (cooperative effects). Indeed, the myriad of defect types and interactions in nuclear fuel under irradiation naturally leads to the supposition that the deleterious losses in thermal conductivity can be mitigated by controlling defect evolution. The Center thus adopts the **vision** that a first-principles understanding of electron and phonon transport addressing the complexity of irradiation-induced defects will provide the necessary tools to control thermal transport in nuclear fuel.

Our vision will be examined from the perspective of two thrusts. The first will tackle phonon mediated thermal transport in advanced oxide fuels (thorium oxide - ThO_2 and thorium/uranium mixed oxide $Th_{1-x}U_xO_2$). The second thrust will emphasize electron and phonon mediated thermal transport in advanced nitride fuels (uranium nitride - UN and thorium nitride - ThN). Both thermal energy transport phenomena contain rich physics that are not well understood and can be investigated using simple systems.

To meet our vision we have defined four research goals that represent significant challenges in the field of thermal transport. These goals are enumerated below:

1. *Extend computational and experimental framework to temperature extremes*

   This will be crucial for developing a fundamental understanding of fuels at operating temperatures. The emphasis will be on fully extending our computational effort beyond 3rd order anharmonicity. To do this we will compare computed and measured phonon dispersion relationships, linewidths, elastic constants, Raman spectra, and thermal conductivities in perfect single crystals of $Th_{1-x}U_xO_2$ measured in extreme environments to reveal the significance of beyond 3rd order anharmonicity.

2. *Accurately measure electron-phonon coupling*

   Electron-phonon coupling is a controlling mechanism in electron-mediated thermal transport. The emphasis will be on using measurement orthogonality (multiple investigative methods spanning different aspects of the solution space) to accurately gauge the impact of electron-phonon coupling. This approach will involve first-principles modeling in tandem with INS/IXS and ARPES to investigate electron-phonon coupling from an electron and phonon perspective in ThN.

3. *Characterize the spectrum of defects and model defect carrier interactions*

   This is critical to developing accurate, first principles informed models of thermal energy transport under irradiation. The emphasis will be on using measurement orthogonality to characterize the size and distribution of sub-nanometer defects (defects that cannot be statistically characterized using

TEM techniques) in oxide and nitride fuels. This will be followed by a first-principles treatment of scattering of energy carriers by irradiation-induced defects.

4. *Understand defect segregation at interfaces and thermal transport across interfaces*

   Defect segregation at interfaces is a prototypic defect evolution mechanism that has been shown to have a net beneficial impact on thermal transport. Additionally, from a thermal transport perspective, interfaces can be studied in isolation using new experimental tools, allowing for measurements to be compared directly to atomistic prediction. Our emphasis will be on naturally occurring grain boundaries in oxide fuels and epitaxially grown heterointerfaces in nitride fuels.

Tackling the computational complexity is a far-reaching challenge. At the atomistic scale, the approach will involve using density functional theory (DFT) and beyond DFT methods able to capture many-body interactions to understand the role of 5*f* electrons on phonon and electron transport, defect formation, and scattering mechanisms. At the mesoscopic-length scale, thermodynamic modeling, molecular dynamics, and kinetic Monte Carlo will be used to understand defect interaction and evolution. Phonon-scattering mechanisms will be investigated using both perturbative and Green's function approaches and their impact on thermal transport will be captured using the Boltzmann transport equation (BTE).

These modeling approaches will be complemented by a well-defined set of electron- and phonon-structure measurements and transport measurements in ion-irradiated model fuels having well-characterized microstructures. Sample synthesis routes include hydrothermal and flux growth of large single crystals as well as heterointerfaces assembled using molecular beam epitaxy. Inelastic neutron and X-ray 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 nitride fuels. Transmission electron microscopy, positron annihilation spectroscopy, and optical spectroscopy will be used to characterize the spectrum of defects produced by ion irradiation. Thermal wave microscopy combined with coherent acoustic wave spectroscopy will be used to make spatially resolved thermal transport measurements across heterointerfaces and isolated grain boundaries as well as the damage plateau in ion-irradiated samples. Scanning transmission electron microscopy and electron energy loss spectroscopy will be used to measure localized, phonon modes associated with interfaces.

Center for Thermal Transport under Irradiation (TETI)	
Idaho National Laboratory	David Hurley (Director), Boopathy Kombaiah, Krzysztof Gofryk, Zilong Hua, Amey Khanolkar, Kaustubh Bawane, Chao Jiang, Linu Malakkal, Shuxiang Zhou, Brelon May, Yanwen Zhang
Oak Ridge National Laboratory	Michael Manley
Airforce Research Laboratory	Mathew Mann
Columbia University	Chris Marianetti
The Ohio State University	Marat Khafizov
University of Texas at San Antonio	Elizabeth Sooby
Bowling Green State University	Farida Selim
University of Wisconsin	Yongfeng Zhang
The Pennsylvania State University	Miaomiao Jin

Contact: David Hurley, TETI Director, david.hurley@inl.gov
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Institute for Quantum Matter (IQM)
EFRC Director: Collin Broholm
Lead Institution: Johns Hopkins University
Class: 2018 – 2024

Mission Statement*: To realize revolutionary quantum materials and structures in which entanglement and coherence drive collective quantum effects.*

Understanding and controlling quantum matter presents one of the grand challenges at the intellectual frontier of the 21st century physical sciences. By enabling new technologies, this fundamental research on crystalline solids has the potential to transform almost every aspect of modern life. As we confront the demands of unprecedented growth in global energy needs and seek to extend the information technology revolution, new materials developed though a deep understanding of the underlying fundamental quantum physics will play an essential role.

The mission of the Institute for Quantum Matter (IQM) is to discover and understand emergent properties in material systems that have the potential for transformative impacts on energy and information technologies. IQM tightly integrates materials discovery and synthesis (single crystals and thin films), advanced experimentation (neutron scattering, optical spectroscopies, transport, ultrasound, tunneling, and high magnetic field techniques) and theory (analytical and numerical).

In the past funding period, IQM discovered new quantum materials and new quantum collective phenomena including magnets with strong quantum fluctuations, superconductivity in a Dirac material with massless charge carriers, and topological materials where quantum transport properties can be controlled by magnetic fields and stress. IQM’s unique inter-disciplinary research environment leads to unique training opportunities for students and postdocs so that IQM alumni have gone on to start their own groups and become quantum science leaders in their own right.

In the present funding period IQM will focus on selected high impact collaborative projects involving multiple PIs and combining a full range of synthesis, spectroscopic, and theoretical methods to achieve conclusive answers to key questions in quantum materials science. The research will be organized in three interlocking thrusts:

Quantum Entangled Magnetism. While conventional magnetic materials contain ordered static structures of atomic scale magnetic moments, these continue to fluctuate in quantum entangled magnets and give rise to unique collective materials properties. Prospective applications may be anticipated in quantum computing and quantum sensing. Building upon discoveries at IQM and elsewhere, IQM will seek evidence for quantum entangled magnetism in single crystalline materials at low temperature and when subjected to extreme conditions of pressure, strain, and magnetic fields. We shall explore quasi-two-dimensional magnets with competing interactions based that promote quantum fluctuations. We shall synthesize ultra-pure single crystalline samples of quantum spin liquid candidates and explore their physical properties, contrasting them with the properties of more disordered samples. Prompted by our recent theoretical and experimental discoveries, we shall examine how strain in 2D quantum magnets affects collective magnetic properties. We shall explore the quantum magnetism of honeycomb spin-1/2 materials based on the magnetically anisotropic $3d^7$ Co^{2+} ions in pursuit of quantum entangled magnetism through high field measurements and chemical doping.

Engineering Correlated Superconductivity. Superconductors have obvious applications in energy distribution and as the basis for qubits in quantum computers, while simultaneously giving new insight about macroscopic quantum physics. Based on our activities in the previous grant period, and recent broader discoveries, we shall explore superconductivity built from correlated, topological, and quantum entangled matter. We shall complete ongoing collaborative IQM projects exploring superconductivity in metals with light electrons resulting from topologically protected band crossings. IQM has developed evidence for anomalous bulk superconductivity in an intermetallic material that may be linked to the Dirac nature of the electronic band structure. IQM will explore the possibility of superconductivity in related materials with magnetic rare earth ions. Using the Josephson effect, we shall explore the properties of proximitized superconductivity in Mn_3X thin films samples sputtered at IQM (see below).

Magnetic Topological Materials and Phenomena. IQM will develop new materials functionality based on electronic topology in conducting and insulating single crystals and thin films that incorporate magnetism. A signature focus is on the Mn_3X family of compounds where we are examining the rich interplay between antiferromagnetic domains and electrical transport including electrical domain switching monitored through the anomalous Hall effect. Proximity induced superconductivity will be explored by sputtering patterned superconducting films onto Mn_3X. We shall use small angle neutron scattering to study the domain wall structure and hysteretic field response of topological magnetic semimetals. These experiments may pave the way to the use of neutrons to probe electrically driven domain switching. Inelastic neutron scattering will be used to determine interactions between rare earth magnetic ions mediated by Weyl fermions. We shall explore magnetic topological insulators with the potential to display strong quantized magneto-electric responses.

Institute for Quantum Matter (IQM)	
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Center for Novel Pathways to Quantum Coherence in Materials (NPQC)

EFRC Director: Joel E. Moore
Lead Institution: Lawrence Berkeley National Laboratory
Class: 2018 – 2024

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.

The research in this Center will dramatically expand our control and understanding of coherence in solids by building on fundamental materials discoveries in recent years. Advances in quantum materials including two-dimensional materials and strongly correlated materials lead to remarkable new kinds of defects, boundaries, and interfaces. Common threads are the need to understand defects in complex material environments, the use of high-resolution optical techniques and other cutting-edge experimental tools, and the role of inhomogeneity in various types of quantum materials.

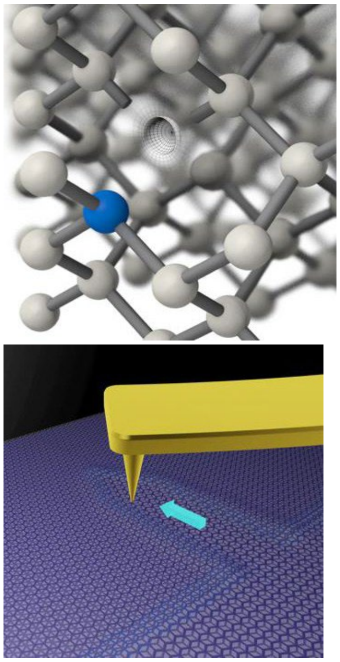

Figure 1. Illustrations of research directions for the thrusts of this EFRC. From top to bottom: a nitrogen-vacancy (NV) center in the diamond lattice used for quantum sensing; controlled writing of one-dimensional boundary in atomically thin 2D materials.

The Center conducts research in two linked major thrust areas. Each thrust involves an integration of synthesis, characterization, and theory, and takes advantage of unique national laboratory capabilities.

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. Harnessing many-body entanglement can significantly enhance such quantum sensing technologies. The goal of this thrust is to theoretically predict, computationally optimize, and experimentally create, characterize, and develop novel defect-based quantum sensing platforms. In addition to the platforms themselves, NPQC investigates 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 nature of correlated states at ultrahigh pressures, and other important materials properties. NPQC research has already demonstrated improved control over the creation and probing of defect ensembles, and begun to apply the improved sensors to important problems in the physics of quantum materials, such as those arising in Thrust 2.

Thrust 2 – Coherent transport and switching in engineered surfaces and layered materials

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. An example of such an interface from previous NPQC work is the creation of Luttinger-liquid behavior at mirror twin boundaries in a 2D dichalcogenide.

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. Superconductivity can emerge for particular arrangements of 2D sheets. Layered crystals made from 2D sheets can use magnetic intercalants to create ordered states that imprint directionality on transport properties in a switchable fashion. New materials in this family 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.

An important benefit of research in NPQC is its role in training junior personnel to become productive scientists in this field at the intersection of quantum materials and quantum information science applications. Further information about NPQC research and activities is provided at the website below.

Center for Novel Pathways to Quantum Coherence in Materials (NPQC)	
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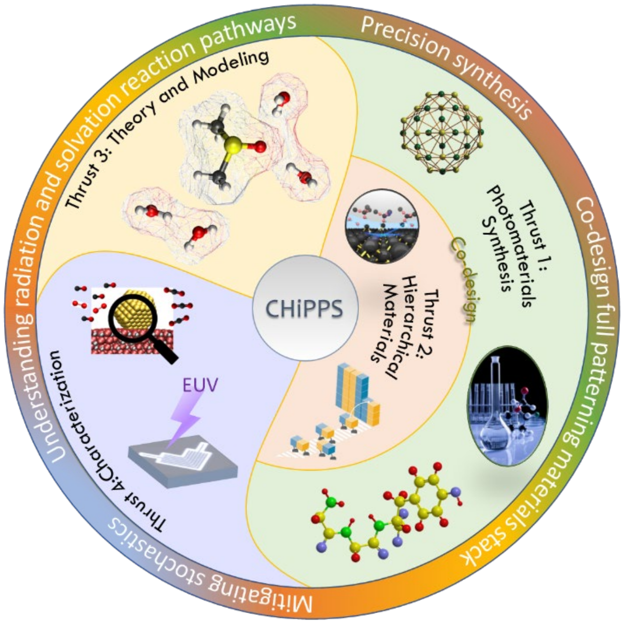
Center for High Precision Patterning Science (CHiPPS)
EFRC Director: Ricardo Ruiz
Lead Institution: Lawrence Berkeley National Laboratory
Class: 2022 – 2026

Mission Statement*: to create new fundamental understanding and control of patterning materials and processes for energy-efficient, large-area patterning with atomic precision, thereby enabling at-scale advanced manufacturing of future generation microelectronics such as quantum and spin-based memory, storage, and logic devices.*

Microelectronics is arguably the single, most critical example of advanced manufacturing, representing a $442B/year economic engine that today drives every sector in the global economy, making it essential to the U.S. economy, and its physical and energy security. Continued scaling of at-scale advanced microelectronics manufacturing to the realm of atomic scale precision is critical to realizing the potential of future generation microelectronics such as quantum and spin-based memory, storage, and logic devices that will in turn enable transformative compute capabilities, while minimizing compute center carbon footprints.



It is evident that continued progress in microelectronics, as well as maintaining U.S. advanced manufacturing leadership, is predicated on future advancements in patterning methods and materials. Even new future technologies that are less dependent on two-dimensional scaling, such as quantum computing and spin-based systems, are also patterning constrained because achieving their promise requires patterning precision well beyond the limitations of current methods and materials.

CHiPPS will pursue its holistic approach to patterning science by studying the fundamentals of relevant interactions of light and matter and proposing co-designed materials for precision patterning from monodisperse or sequence defined resists to molecular-level control of solvation steps to bottom-up hierarchical and functionalized materials for low-impact stochastics to molecularly precise and selective pattern transfer. Our research comprises four highly integrated thrusts to meet the Mission CHiPPS:

1. **Thrust 1. Patterning Materials.**
   This thrust is focused on the design and synthesis of top-down radiation sensitive materials to control reaction fronts and stochastic variation in chemical patterning at sub-nm length scales.
2. **Thrust 2. Bottom-up hierarchical materials and processes.**
   Thrust 2 will further integrate bottom-up hierarchical materials and processes with Thrust 1 materials to *heal* the adverse stochastic effects on the photoresists and to enable pattern transfer methods with atomic or molecular precision. Thrusts 1 and 2 will be tightly integrated and coordinated within CHiPPS, enabling co-design of top-down and bottom-up materials and processes.

3. **Thrust 3. Theory and Modeling.**
   Thrust 3 will guide thrusts 1 and 2 by implementing quantitative theory and modeling for rational design of patterning materials. The modeling will also be informed by thrust 1 model materials.
4. **Thrust 4. Multimodal Characterization.**
   The cross-cutting thrust 4 will develop unique operando characterization providing critical feedback and validation of design rules for Thrusts 1 and 2 as well as validating Thrust 3 theory. Thrust 4 will include at-scale advanced manufacturing relevant characterization by leveraging the world's most advanced EUV patterning facility built at the Advanced Light Source.

Center for High Precision Patterning Science (CHiPPS)	
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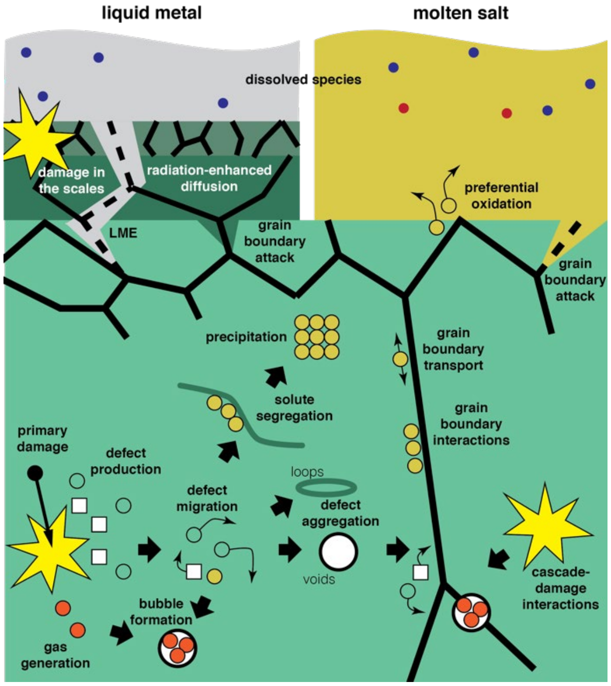
Fundamental Understanding of Transport Under Reactor Extremes (FUTURE)
EFRC Director: Blas Pedro Uberuaga
Lead Institution: Los Alamos National Laboratory
Class: 2018 – 2026

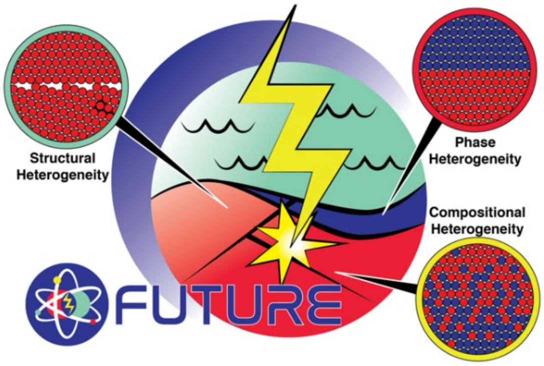
***Mission Statement**: To understand how the coupled extremes of irradiation and corrosion work in synergy to modify the evolution of materials by coupling experiments and modeling that target fundamental mechanisms.*

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 that comprise the system. These extremes include irradiation, temperature, stress, and corrosion. Individually, irradiation damage and corrosion are two of the greatest materials science challenges as they are truly multiscale. For example, radiation effects span 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. Corrosion mechanisms differ depending on if the corrosive environment is a molten salt versus an oxygenated system that induces oxide formation. Coupling irradiation with corrosion leads to an immense scientific challenge requiring a multidisciplinary team, just as we have assembled such a team in FUTURE.



In FUTURE, we target the response of materials to a combined corrosive and irradiation environment. 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. Building on the success of the first phase of FUTURE, in this second phase we target materials heterogeneities. Real materials are characterized by heterogeneities – “disruptions of perfect order” – that often dictate their response to extreme conditions. These include structural heterogeneities such as grain boundaries and dislocations, phase heterogeneities in which multiple phases are present at once, and compositional heterogeneities where chemical species are distributed non-uniformly throughout the material. At the



same time, radiation damage and corrosion both induce new heterogeneities in the material, leading to a dynamic feedback between the response and structure. In FUTURE, we focus on these heterogeneities with the goal of understanding how they couple to radiation damage evolution and corrosion mechanisms to modify the response of the material to these combined extreme environments. This coupling leads to three hypotheses that guide the scientific research of FUTURE:

- The energy landscape for transport in compositionally-heterogeneous alloys and oxides alters the rate and prevailing mechanisms of corrosion and is, in turn, modified by irradiation.
- The thermokinetics of defect evolution and thus ongoing corrosion, thermally and under irradiation, differ in a multiphase vs. single phase material.
- The dynamics of transport that drive corrosion through extended defects and their networks are altered under irradiation.

Understanding how these heterogeneities impact the coupling between irradiation and corrosion requires a wide-range of expertise in radiation damage and corrosion science, unique modeling techniques, and integrated experimental facilities that can only come from a Center such as FUTURE. We have developed novel modeling and experimental capabilities to interrogate and understand this coupling. These include in-situ positron annihilation spectroscopy, where the positron beam is coincident with an ion beam to probe damage evolution in situ; the Irradiation-Corrosion Experiment, which provides the ability to subject a material to irradiation while also be exposed to a corrosive environment; buried isotope markers and atom probe tomography, to directly quantify radiation-enhanced diffusion; a cluster dynamics model of coupled irradiation and corrosion, accounting for the myriad of reactions that describe the interaction between radiation-induced defects and corrosion mechanisms; and a new multi-phase field model that predicts the morphological evolution of metallic alloys in a dealloying regime.

The goal of FUTURE is to reveal the fundamental factors dictating the evolution of materials under the coupled extremes of irradiation and corrosion with a goal of developing a predictive capability for these materials.

Fundamental Understanding of Transport Under Reactor Extremes (FUTURE)	
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Pacific Northwest National Laboratory	Danny Edwards, Tiffany Kaspar, Dan Schreiber, Steven Spurgeon, Sandra Taylor, Kayla Yano
Arizona State University	Farida Selim
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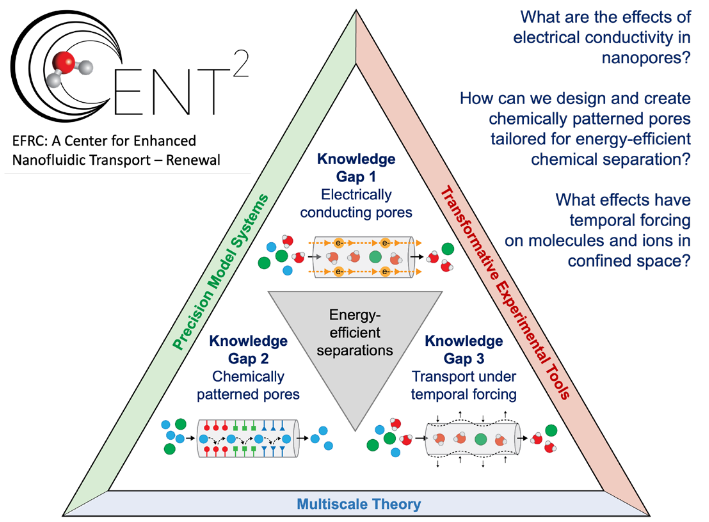
Center for Enhanced Nanofluidic Transport Phase 2 (CENT2)
EFRC Director: Michael S. Strano
Lead Institution: Massachusetts Institute of Technology
Class: 2018 – 2026

Mission Statement*: CENT2 is a pioneering Energy Frontier Research Center addressing critical knowledge gaps in our understanding of fluidic flow and molecular transport in extremely narrow pores. CENT2 will establish the scientific foundation for transformative molecular separation technologies impacting the water-energy nexus and Energy Earthshots.*

Not all nanopores are created equal. By definition, all have characteristic diameters or conduit widths of between approximately 1 and 100 nm. However, the narrowest of such pores, **S**ingle **D**igit **N**anopores (SDNs), are less than 10 nm in diameter and have only recently become accessible for precision transport measurements. What scientists within the Center for Enhanced Nanofluidic Transport (CENT) and the larger community have discovered about SDNs has been surprising, including demonstrations of extra-ordinary molecular flux and selectivities beyond theoretical predictions. These findings have in turn exposed critical gaps in our understanding of hydrodynamics, molecular sieving, fluidic structure, and thermodynamics under the confinement of SDNs that form the basis of CENT. Building on our groundbreaking work, the renewed CENT (**CENT^2**) will address fundamentally new questions of fluidic transport at the nanoscale that stand to impact a host of new technologies at the water-energy nexus, from new membranes for precise separations and water purification, to new permeable materials for water electrolyzers and energy storage devices, as well as enabling new emerging energy technologies such as hydrovoltaics. Our effort directly responds to the need for *Basic Research in Energy and Water* as outlined in the DOE-EFRC FOA. Furthermore, the fundamental knowledge gained on the selective transport of molecules and ions as part of CENT^2 will broadly support the *Energy Earthshot* cross-DOE initiatives.



CENT^2 is organized into *three* distinct, cross-cutting knowledge gaps to establish the scientific foundation for transformative molecular separation technologies.

CENT^2 will continue as a pioneering Energy Frontier Research Center, leveraging its newfound insights of fluidic flow and molecular transport in extremely narrow nanopores to inspire the next generation of water purification and membrane separation processes. This vision encompasses vastly more precise and energy efficient separations at the water-energy nexus as well as in many other sectors of the U.S. economy. In phase 2, our Center will focus on *three* distinct and interconnected Knowledge Gap (KG) activities to advance our four-year research goals: **Electrically Conducting Pores (KG1); Chemically Patterned Pores (KG2); and Transport under Temporal Forcing (KG3).** CENT^1 has developed fundamental understanding of **slip flow enhancement** (i.e., substantially higher mass transport rates in *smaller* nanopores), which now enables us to devise new mechanisms of fluidic enhancement, such as electrically driven boundary flow, involving the coupled transport of fluid in the SDN and electrons in the confining material that forms the nanopore (**KG1**). Our thermodynamic measurements inside SDNs have uncovered fluid phase boundaries that are remarkably distorted from their bulk fluid counterparts, demonstrating the so-called **non-Gibbs-Thomson** behavior and leading to an inability of existing models to predict the existence of vapor, liquid or solid phases within the narrowest of pores. **KG2** then seeks to use chemically patterned SDNs that are only otherwise found in nature, to manipulate fluid phase boundaries as well as transport behavior at the extreme confinement for fundamentally new separation regimes. Finally, **KG3** will examine molecular transport and separations under forced modulation, either electrokinetic (**3A**) or mechanical (**3B**), with tunable frequency and amplitude to exert new types of control over transport at the molecular scale. **These fundamental studies hold the potential to uncover new insights for the development of next-generation water purification and membrane separation processes that could encompass self-pumping membranes, pores with strongly defined and tunable single-species selectivity, and feedback control schemes that directly modulate the transport medium itself.** We define these three new areas of investigation as **knowledge gaps** because existing theories, whether continuum, atomistic, or molecular simulations, fail to adequately quantify or even describe the basics of these exotic effects. Leveraging the knowledge gained from the previous performance period, CENT^2 will establish the foundation for transformative molecular separation technologies impacting basic energy research needs. We have assembled a new multidisciplinary team of the leading experts in SDN research from MIT, UMD, Yale, UCI, Stanford, Univ. Florida, and LLNL, including the addition of a Hispanic serving institution, UT Austin, and the addition of three faculty PIs that further expand our diversity. The basic science developed by CENT^2 will enable new energy-efficient separation processes. We address the DOE *Grand Challenge* of the atomic-level synthesis of new materials that exhibit precise control of molecular permeation. Additionally, the fundamental knowledge gained by CENT^2 on the selective transport of molecules and ions will be foundational to addressing the *Energy Earthshots*.

Center for Enhanced Nanofluidic Transport Phase 2 (CENT2)	
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University of California Irvine	Zuzanna Siwy, Javier Sanchez-Yamagishi
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Stanford University	Arun Majumdar
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The Center for Catalysis in Biomimetic Confinement (CCBC)
EFRC Director: Cheryl Kerfeld
Lead Institution: Michigan State University
Class: 2022 – 2026

Mission Statement*: To understand the means by which Nature spatially organizes catalysis across scales using compartmentalization within selectively permeable protein-based membranes, and to use these principles to develop a modular platform for spatially organizing catalysis.*

Biology accomplishes remarkable N_2 and CO_2 fixation reaction pathways in complex environments at ambient temperature and pressure by exploiting confinement effects in charge-accumulating architectures. Confinement features range in scale from the entatic control of catalysis within the enzyme active site to the organized encapsulation of multi-step reaction sequences within subcellular organelles. While organelles have traditionally been considered the defining feature of higher (eukaryotic) organisms, recently it has become clear that bacteria also contain metabolic organelles, known as Bacterial Microcompartments (BMCs)[1, 2]. BMCs function analogously to eukaryotic organelles like the chloroplast; by spatially sequestering sequential enzymes within a selectively permeable membrane (the shell), BMCs co-confine catalysts, establish spatial control of local reactant and substrate concentrations, sequester volatile or reactive intermediates, separate competing/incompatible pathways, and insulate catalysts from inhibitors. The most extensively characterized BMC, the carboxysome, confines ribulose-1,5-bisphosphate carboxylase-oxygenase (RuBisCO) and carbonic anhydrase (CA) in a selectively permeable membrane (**Figure 1**) that enables concentrating CO_2 while simultaneously providing a barrier to O_2, RuBisCO's competitive inhibitor.

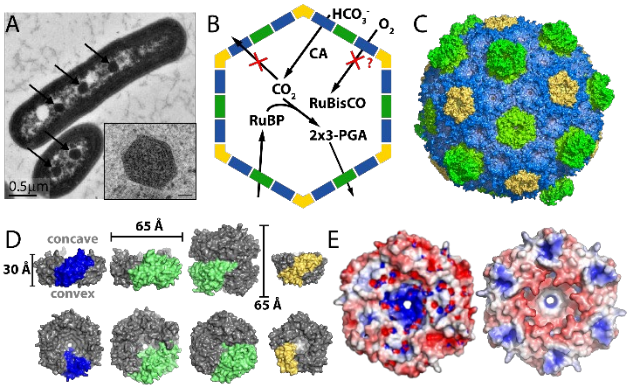
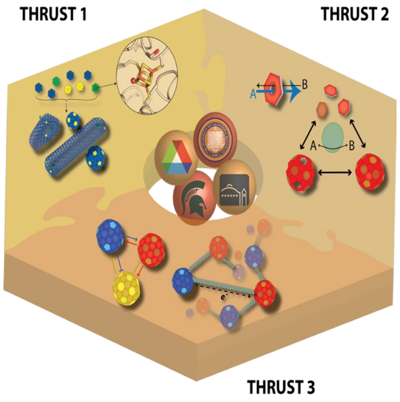


Figure 1. Structure, function, and context of BMCs. A) Cyanobacteria with carboxysomes (arrows); inset, close up view of a 200 nm carboxysome. B) Schematic of carboxysome function. C) Structure of a 6.5 MDa BMC shell determined by a combination of X-ray crystallography and Cryo-EM[3]. D) Basic building blocks of all BMC shells. E) Shell protein hexamers colored by electrostatic potential; note distinct charge around central pores of 4 and 7Å, respectively.

BMCs have evolved to naturally overcome challenges associated with the enormous complexity of metabolic pathways by confining reactions that are incompatible with the surrounding environment and by sequestering pathways to prevent unwanted metabolite crosstalk. The modularity of shell proteins and their potential for self-assembly in both heterologous expression systems and in vitro presents a remarkable opportunity to understand shell protein properties and the principles of self-assembly, as well as to develop a modular tunable framework for spatially confined chemistry. Unlike lipid-bound compartments, the protein-based membrane of the BMC can be precisely structurally defined, and the multiple shell constituents can be individually tuned for electron, substrate, product, and gas transport properties, and provide spatially controlled recognition/anchoring sites on the shell's interior and exterior surfaces for catalysts. BMCs are unique among both biological (liposomes) and synthetic (e.g., zeolites, MOFs, and mesoporous materials) compartments because their shells are essentially a chemically active material that can be tuned or evolved for site-specific/task-specific function using gene-based engineering techniques.

The CCBC is focused on elucidating the underlying principles of BMC structure and function to enable extending and repurposing shell protein building blocks as modular platforms for structuring catalysis at scales extending from the catalyst active site to hierarchical organized micron-scale assemblies of catalytically and redox-active shells. Integral to this goal is understanding the biophysical and mechanistic impact of confinement on enzymes and abiotic catalysts. The CCBC leverages National Lab facilities and bring together a multidisciplinary team comprised of structural and synthetic biologists, enzymologists, spectroscopists, computational theorists, photochemists, and inorganic and synthetic chemists to create an integrated effort addressing fundamental questions on hierarchical confinement for the control of complex, energy-transducing reaction networks. This multidisciplinary team will share a well-developed model shell protein platform combined with computational modeling and selected enzymatic and synthetic catalysts to elucidate the underlying physical principles utilized by Nature in its extraordinary variation of BMC function, thereby enabling us to apply those principles to develop new catalytic systems. This work is groundbreaking by employing BMCs as a tunable, building-block compartmentalization systems for bio-hybrid synthesis. The integration of synthetic biology, chemistry, and catalysis enables hierarchical confinement to be investigated as a general physical-chemical concept applicable for controlling non-natural biological and synthetic energy-conserving chemistries. This will be investigated at unprecedented levels of scale by using BMCs as tunable nano-reactors that can be connected into 2D and 3D hierarchical networks.



The CCBC includes a utilization of natural and synthetic catalysts, protein-based compartments, and both in vitro and in vivo assembly approaches to provide a fundamental, cross-cutting mechanistic understanding for engineering and control of catalysis in confinement. The CCBC is organized in three linked Thrusts (**Figure 2**) representing progressively increasing scales of organization. Thrust 1 focuses on characterization of assembly and microenvironments within BMC shells; Thrust 2 uses BMCs to investigate confinement effects n energy-converting chemistries; Thrust 3 investigates the mesoscale assembly and organization of compartmentalized catalysis.

Center for Catalysis in Biomimetic Confinement (CCBC)	
Michigan State University	Cheryl Kerfeld, Eric Hegg, Dan Ducat, Michael Feig, Robert Hausinger, Markus Sutter, Michaela TerAvest, Josh Vermaas
Argonne National Laboratory	David Tiede, Karen Mulfort, Oleg Poluektov, Lisa Utschig
Berkeley National Laboratory	Corie Ralston

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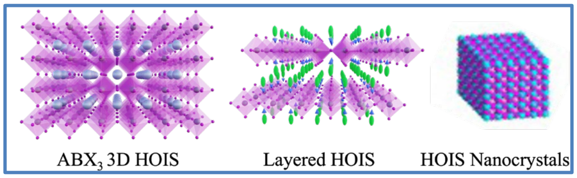
Center for Hybrid Organic Inorganic Semiconductors for Energy (CHOISE)
EFRC Director: Matthew C. Beard
Lead Institution: NREL
Class: 2018 – 2026

***Mission Statement**: To demonstrate unprecedented control over spin, charge, phonon and light properties through synthesis and characterization of crystalline Hybrid Organic Inorganic Systems, their interfaces and heterostructures.*

Hybrid organic-inorganic semiconductors (HOIS) exhibit unique physical phenomena resulting from synergistic interactions between organic and inorganic sub-components. The central hypothesis of CHOISE is: ***HOIS offer unprecedented opportunities to control fundamental physical properties for the next generation of sustainable energy technologies.*** CHOISE researchers are validating this hypothesis by addressing fundamental knowledge gaps of organic-inorganic interactions in this new class of 'unconventional' semiconductors. While *unprecedented* world-wide progress towards efficient HOIS PV systems argues for their unique physio-chemical properties, we have just scratched the surface of their full potential, and basic scientific understanding still lags far behind demonstrated results. CHOISE research aims to broaden HOIS by elucidating factors that control and define their exceptional properties. CHOISE proposes to focus on: precise synthetic pathways leading to well-defined heterostructures; understanding and controlling ion migration; controlling strain; chiral and other symmetry breaking motifs; magnetic ordering and quasi-2D magnetic systems; and impact of the soft lattice on defects, doping, and physio-chemical properties.



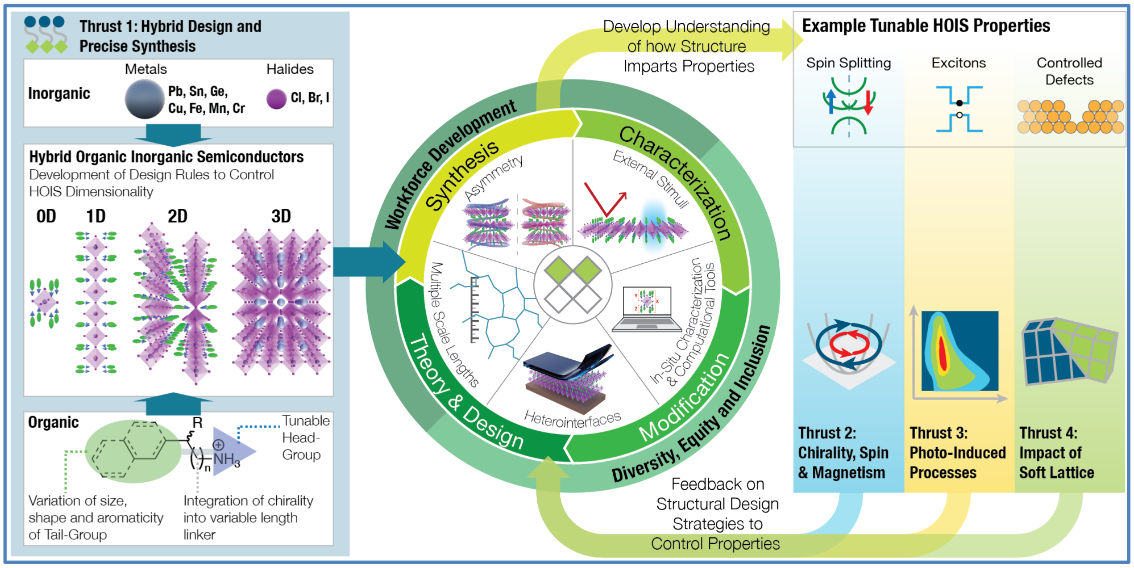
HOIS are unconventional semiconductors with properties that vary continuously from those associated with organic semiconductors having tightly bound Frenkel-type excitons characterized by large exciton binding energies, to those associated with inorganic semiconductors exhibiting delocalized Wannier-type excitons and free-charge carriers. However, the most intriguing aspects of HOIS are emergent properties, i.e., those that neither exist in organic or inorganic systems *but arise due to the interplay between the subcomponents.*

A prototypical HOIS has perovskite structure with ABX_3 stoichiometry where A is an organic cation such as methylammonium ($CH_3NH_3^+$), B is a metal cation such as Pb, and X is a halide anion. CHOISE efforts in 3D HOIS relate to understanding defects, strain, ion-migration, and controlling carrier dynamics. A broader class of hybrid compounds exist with HOIS in non-perovskite structures, low-dimensional structures, or with non-typical A/B/X species. Low-dimensional HOIS structures form when the organic cation is too large and/or an additional secondary organic molecule is added. HOIS nanocrystals (NCs) (e.g., quantum dots, nanoplatlets, nanorods, nanowires) provide unique opportunities via larger tolerance of A-site cations and influence from organic surface ligands.

Spin-splitting occurs in the presence of spin-orbit coupling (SOC) and inversion asymmetry that locks the spin character to the linear momentum and creates unique spin textures. Incorporating chiral organic cations with the inorganic subsystem results in chiral-HOIS. Electrons transmitted through such oriented chiral systems become spin-polarized. This effect of chirality-induced spin selectivity (CISS) can impact a wide range of potential applications such as low-power electronics, chiral-light detectors and generators, electrochemical water splitting, enantiomer separations, and potentially many others not yet envisioned.

The soft nature is a distinguishing property setting HOIS apart from semiconductors like Si and GaAs. Softness reflects dynamic disorder, where ions undergo large and strongly coupled motions about their average lattice positions. The soft nature arises from weak noncovalent bonding and coupling between

lone pair *s* orbitals of the B cation and halide *p* orbitals. Weak bonds that are relatively easy to tilt, vibrate, and break significantly influence structural, electronic, optical, and defect properties, leading to dynamic behavior, ferroelasticity, ferroelectricity, electrostriction, hot-carriers, large polarons, broadband light emission, low recombination rate, and the challenge to extrinsically *n*- or *p*-type dope HOIS.
CHOISE goals are:



Inorganic/organic components form crystalline HOIS wherein the inorganic component may comprise 0D isolated anions, 1D chains, 2D/quasi-2D planes, or completely interconnected 3D network. CHOISE is uncovering the basic understanding and design rules to uncover unprecedented control over spin, light and charge.

- Develop precise synthetic methods allowing for atomic, micro-, and macro-level control over composition, structural dimensionality, symmetry breaking, phase instability, heterostructures, and defects.
- Uncover the mechanism(s) of spin-selectivity, develop control over spin-textures, and induce novel quasi-2D magnetic behavior by manipulating symmetry breaking motifs.
- Understand the interconversions between light, excitons, spins, and charge carriers, gaining control over photocatalytic transformations, hot-carrier utilization, spin-transfer, and polarized light absorption/emission.
- Uncover impacts of micro- and macro-scale strain, doping, lattice dynamics, ion migration, and defect distributions and feedback to enable synthesis of novel HOIS.
-

Center for Hybrid Organic Inorganic Semiconductors for Energy (CHOISE)	
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National Renewable Energy Laboratory	Matthew C Beard, Joseph Luther, Kai Zhu, Jeffrey Blackburn, Peter Sercel
University of Wisconsin-Madison	Yuan Ping
University of Utah	Valy Vardeny
University of North Carolina, Chapel Hill	Jinsong Huang
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University of Colorado Boulder	Mike Toney, Joseph Berry, Seth Marder
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North Carolina State University	Dali Sun

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Center for Electrochemical Dynamics and Reactions on Surfaces (CEDARS)
EFRC Director: Dhananjay Kumar
Lead Institution: North Carolina Agricultural and Technical State University
Class: 2022-2026

Mission Statement: *To reveal the formation of the transient intermediates of oxygen evolution reaction and hydrogen evolution reaction and how the catalyst evolves before, during, and after catalysis.*

Hydrogen produced from water electrolysis is abundant, clean, and secure. This method of producing hydrogen, however, is slow and requires energy beyond what is thermodynamically necessary. At present, the molecular factor(s) responsible for these limitations is not well known because the two electrochemical reactions of water splitting, the oxygen evolution reaction (OER) and hydrogen evolution reaction (HER), are complex cascades of electron-driven reactions with transient species forming and disappearing at each step along the catalytic process. Starting from the descriptions of proton transfer (PT) and electron transfer (ET) to surface bond formation and dissociation, the mission of this center is to reveal step-by-step the formation of the transient intermediates, what causes the chemical bonds to form, how a catalyst evolves before, during, and after catalysis, as a function of time and reaction conditions. A team with the capabilities to deconstruct the mechanism of OER and HER into the elementary steps has been put together to address this challenge. The intended work integrates high-precision growth of materials with studies of the short-lived transient intermediates by optical and X-ray scattering and spectroscopy, and first-principles modeling inclusive of beyond density functional theory (DFT) and grand canonical calculations. The plan requires a team interweaving across disciplines, from materials, chemical, to computational sciences.

Our team integrates investigators from universities and DOE laboratories (**Fig. 1**). The research is organized into three thrusts by the time and event sequence of the catalytic cycle. The research in Thrust 1 focused on determining how the water dissociation energies and dynamics on oxide catalysts containing 3d, 4d, and 5d metals. Identifying the electrochemical dynamics of the metal-oxygen bond as a function of electronic structure, surface composition, and excitation potential will reveal how the starting point of an electrochemical reaction varies between different catalysts. CEDARS will focus on metals, metal oxides, and oxynitrides prepared as single-crystal, epitaxial, or conformal films on photo-adsorbers that are oxides or III-V semiconductors. To resolve the molecular structure and formation dynamics of the intermediates, CEDARS will apply multi-modal, *in situ*, and time-resolved spectroscopy. Ultrafast optical spectroscopy and ultrafast X-ray absorption spectroscopy will in-tandem assign electronic orbitals to the optical transitions of charge-transfer intermediates and derive changes in the covalency and metal-oxygen bond distance.

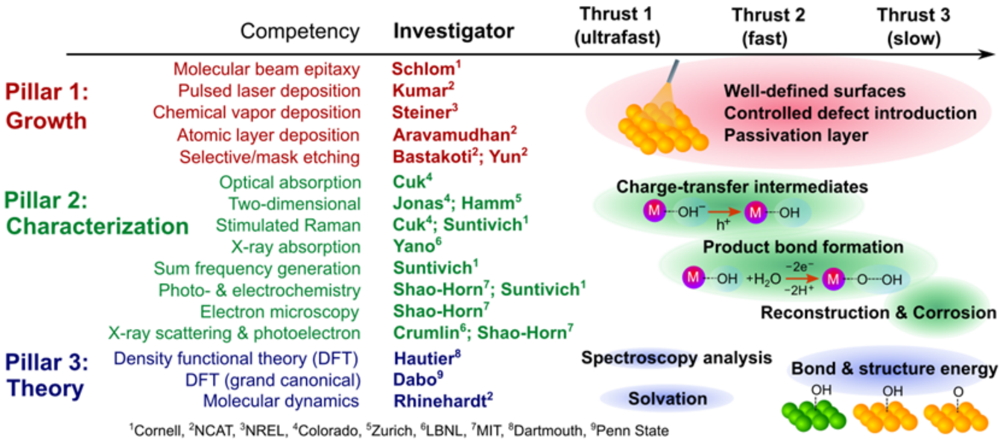

Fig.1: CEDARS team and intertwined three research thrusts.

Thrust 2 is concerned with resolving how the reaction kinetics are related to surface thermodynamics and the local environment of electric fields, diffusive motion of charge and ions, and chemical exchange, controllable by interfacial chemistry and device structure. The challenge implicit to this Thrust is the need to probe the interface holistically to find the reaction coordinate. CEDARS will harness two strategies: (a) Systematic tuning of the interfacial environment through reaction chemistries, spatial control, and select-area excitation and (b) Employing multi-modal and multi-dimensional probes to follow interacting intermediates through bond formation, utilizing the full electromagnetic spectrum and non-linear spectroscopy. The team will use these approaches in tandem to reveal surface dynamics related to bond formation. The approach and findings of Thrust 2 will allow the team to interrogate how the electronic and chemical states favor specific reaction pathways and how to tailor them for faster bond formation for better catalysis.

The research in Thrust 3 is focused on establishing a connection between catalytic turnover, surface re-structuring, and stability. CEDARS will detect how the underlying materials adapt during the formation of transient intermediates and the accompanying chemical bonds building on the intermediates and bond formation events that occur at a fast-time scale (> 10^{-6} s). The surface changes that arise from M-O, O-O, and H-H bond distortions are small but can cumulatively initiate failure event such as corrosion. CEDARS will explore two strategies to limit the corrosion: (1) Applications of thin passivation layers to allow the subsurface electronic structure to interact with the reactive intermediate but sufficiently inert such that corrosion is suppressed (2) Use the time constants associated with the intermediates that lead to corrosion to inform alternative strategies for engineering stability, such as timing surface activation by electron flow. Comparing the electrochemical and spectroscopic studies from the fast-time domain through many catalytic cycles will allow CEDARS to holistically investigate surface reconstruction and its connection to corrosion. As detailed earlier, *operando* characterization will be performed and alongside online mass spectroscopy. High-throughput DFT calculations will be applied to survey the large space of possible compositions and structures to map possible local motifs of corrosion to the experimental spectroscopic signatures.

The research efforts are complemented by activities in Workforce development, Diversity, Equity & Inclusion, and training. The integrated research and education plan will allow CEDARS to imbue the next-generation workforce with a unique skillset to drive the effort of clean hydrogen production at terawatt scales through multidisciplinary, diverse research collaboration and exchange.

Center for Electrochemical Dynamics and Reactions on Surfaces (CEDARS)	
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Dartmouth College	Geoffroy Hautier
Lawrence Berkeley National Laboratory	Ethan Crumblin, Junko Yano
Massachusetts Institute of Technology	Yang Shao-Horn
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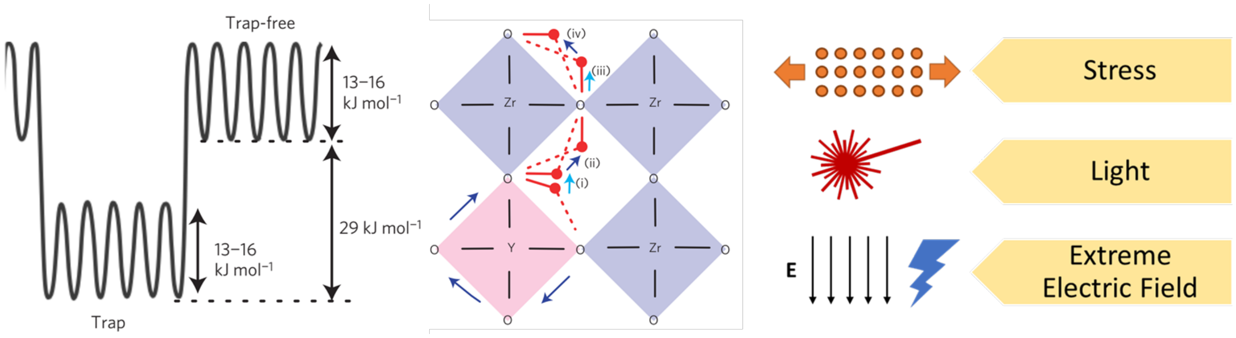
Hydrogen for Energy and Information Sciences (HEISs)
EFRC Director: Sossina M. Haile
Lead Institution: Northwestern University
Class: 2022 – 2026

Mission Statement*: To advance the fundamental understanding and discovery of multihued hydrogen transport in inorganic solids of earth-abundant elements, and of its transfer along and across interfaces within such materials, where 'hydrogen' includes all charge states of the element: H^+ (proton), H^0 (atom), and H^- (hydride ion).*

The transport characteristics of hydrogen are distinct from those of other elements owing to its small mass, which is comparable to the electronic effective mass of some heavy-Fermion solids, and its ambipolar nature. Dominant transport via tunneling at ambient temperatures is possible for protons, with appreciable kinetic isotope effects. Its redox flexibility, occurring as either a cation or an anion and potentially transforming between states within a given host material, is accompanied by a dramatic change in ionic/atomic radius, from effectively 0 in H^+, to 0.6 Å in H^0, and 1.1 Å in H^-. Furthermore, the hydride ion is far more polarizable than other anions of comparable ionic radius, O^{2-} and F^-, suggesting a route to tailoring bond covalency and enabling high mobility.

Recognizing the unique characteristics of hydrogenic species transport, HEISs proposes to exploit external static and dynamic electric/strain fields to **design** the energy landscapes for mobile and reactive H-containing species in bulk crystals as well as at interfaces. Of particular interest and value are (i) exceptionally **high electric fields**, (ii) **high mechanical stresses**, and (iii) **photon** illumination, where the latter additionally induces **phonon** modulation. These external drivers couple to the energy landscape of hydrogen defects and can alter the dominant charge state of H and distribution within trap sites, can serve as a means of engineering higher dimensional defects, and can also drive transport and reaction dynamics at strongly nonlinear field-strength regimes.

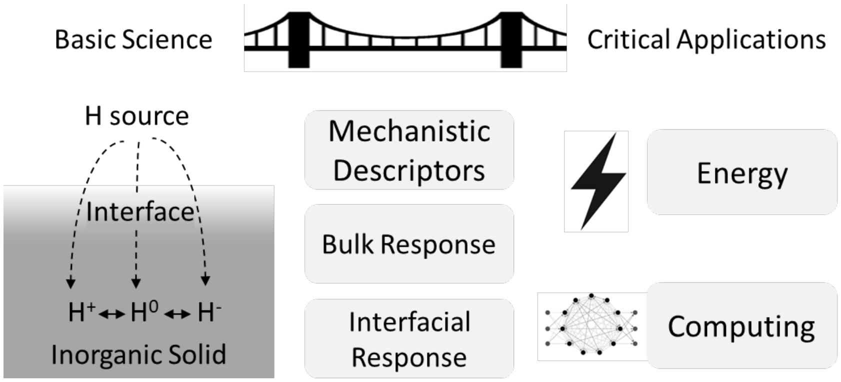


Beyond its special place in physical chemistry, hydrogen is of tremendous societal importance in energy technologies and of growing importance in energy-efficient computing. In both arenas, the relevant devices are limited by hydrogen kinetics, whether it be electrochemical reaction at an interface or diffusion through the bulk, and whether the material be an electrolyte, a semiconductor, or a metal. HEISs will establish the governing mechanisms and physical descriptors of the transport and interfacial incorporation mechanisms needed to achieve precision-guided discovery and design across these classes of materials. In particular, HEISs will address the following critical open questions:

- What are the rate-limiting steps in hydrogen transport and incorporation mechanisms, and the electronic, structural, and dynamic descriptors thereof?

- Can we vary the hydrogen oxidation state within a given material? Is the oxidation state fixed at the point of incorporation, or can it be dynamically tuned?
- What is the nature of traps that inevitably display affinity for hydrogen species? Traps typically retard bulk transport, but can extended defects serve as high mobility highways? And can surface trap sites be designed to achieve desirable surface incorporation pathways?
- Can we exploit stimuli beyond heat, such as pressure, light, and extreme electric field, to accelerate hydrogen dynamics?
- What underlying transport principles apply across multiple classes of oxide materials (wide band-gap electrolytes, small band-gap semiconductors, metallic conductors), and which transport features are unique to a particular class?

With a deliberate focus on use-inspired, ambient-to-intermediate temperatures, the scientific advances and insights gained will provide a foundation for controlling electrochemical transformations critical for carbon-neutral energy (including nitrogen and carbon dioxide reduction) and for modulating electron transport in materials for brain-inspired computing.



Hydrogen for Energy and Information Sciences (HEISs)	
Northwestern University	Sossina Haile (*Director*), Christopher Wolverton (Deputy Director), Michael Bedzyk (DEI Officer), Lin Chen (Liaison to Student/Postdoc Council; Argonne National Lab), Vinayak Dravid, James Rondinelli
Massachusetts Institute of Technology	Ju Li (Theme 3 Lead), Bilge Yildiz
Colorado School of Mines	Ryan O'Hayre (Theme 1 Lead)
University of Illinois, Urbana-Champaign	Nicola Perry (Theme 2 Lead)
Florida State University	Yan-Yan Hu
University of Tennessee, Knoxville	Katharine Page

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Center for Bio-Inspired Energy Science (CBES)
EFRC Director: Samuel I. Stupp
Lead Institution: Northwestern University
Class: 2009 – 2024

***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 conversion of light to chemical energy. Our proposed research program specifically tackles the next big challenges in synthetic design of soft materials, namely learning how to molecularly encode in them the ability to transduce energy 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 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 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. Our cross-disciplinary investigations focus on bio-inspired research in the following areas.

Task 1: "Multi-Scale Synthesis of Artificial Muscles"

Building on computational and experimental design of light-responsive hydrogel walkers and photoresponsive heterogeneous origami gels developed in CBES during the last funding cycle, we will explore new light-responsive hydrogels containing supramolecular polymers that undergo more complex responsive motion, such as swimming toward a light source. Our continuum models are being used to describe the swimming mechanism and the possible origin of phototactic behavior. We will continue to optimize the performance of the swimmer and we will perform simulations for different conformations and robot designs capable of performing various shape transformations for targeted functions. The modeling will include the development of time-dependent, light-responsive hydrogel models that will incorporate multiple reversible transformation mechanisms for robots.

Task 2: "Magnetic Morphogenesis"

CBES researchers have studied the equation of state that arises in a system of colloidal rotors driven by a magnetic field on an inclined surface, which shows unusual behavior with changes in viscosity and frequency of rotation. It has become apparent that we need to include hydrodynamic codes developed by CBES to understand the phenomenon. For example, we are now considering "odd viscosity", which arises if time reversal is broken, in Stokesian dynamics of bulk systems.

Task 3: "Autonomous Soft Microrobots"

During the last funding period, CBES developed Quincke oscillators obtained from micron-scale colloids in a static electric based on the charging and actuation of particles within a field-induced boundary layer. The Quincke oscillation mechanism provides a useful experimental model for active matter composed of many self-oscillating units where particle interactions depend on oscillator orientation and phase to mediate their collective dynamics. Experiments on ensembles of Quincke oscillators have recently revealed new emergent assemblies such as oscillating vortex crystals, where oscillators synchronize and orient within close-packed lattices. We are developing a dynamic model of multiple Quincke oscillations to understand the synchronization and alignment of two oscillators mediated by electrostatic and hydrodynamic interactions. We have also developed active colloids and other soft-matter systems that exhibit internal control mechanisms for achieving autonomous functions like gradient-driven navigation. We are developing a highly dynamic coacervate system based on enzymes in a chemostatted chamber to enable the experimental study of liquid droplets with internal feedback mechanisms based on chemical fuels. In this system, the chemostatted environment promotes drop formation, while the enzymatic reaction within the droplet promotes its dissolution to create dynamic droplets of a prescribed size.

Task 4 "Hierarchical Structure-Mediated Photocatalysis"

As part of our previous studies on non-equilibrium phenomena in hydrogels, we discovered colloidal crystal metallization in binary crystals of size-asymmetric DNA-functionalized nanoparticles with complementary end nucleotide sequences. At the metallization transition, the small component lattice melts. We are now analyzing the dynamic behavior of size-asymmetric charged colloidal particles under an external electric field when the large nanoparticles are connected by polymeric springs. Preliminary work suggests a dynamic transition to a state where the small particles condense into clusters under high electric fields. This work will demonstrate a new way of transporting charge that will be useful for the design of devices with controlled transport, including batteries.

Task 5: "Mechanical Enhancement of Photocatalysis"

Movement in photosynthetic organisms regulates a series of processes that are necessary to maximize light absorption, control gas exchange, and regulate water loss, among other functions. We recently discovered a novel self-assembled system that photocatalytically produces hydrogen peroxide without the need for a metal catalyst. By integrating this supramolecular polymer into a thermoresponsive polymer scaffold, we found that the photocatalytic reaction rates can be controlled by changing temperature. With heating and cooling cycles, the photocatalysis rate increases with the actuation rates due to enhanced flow of water and solutes in and out of the hydrogel. The demonstration that actuation can regulate and even enhance the photocatalytic rates provides a framework for designing future lifelike artificial photosynthetic systems.

Center for Bio-Inspired Energy Science (CBES)	
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Harvard University	Joanna Aizenberg, George Whitesides
Massachusetts Institute of Technology	Danna Freedman
New York University	Paul Chaikin
University of Michigan	Sharon Glotzer
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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.

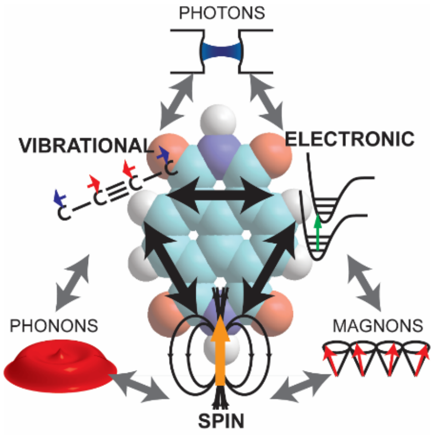

Fig. 1. Synergistic **CMQT** research directions. Thrust 1 focuses mainly on spin and electronic DOFs. Thrust 2 adds phononic and magnonic DOFs. Thrust 3 adds photonic and vibrational DOFs.

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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Massachusetts Institute of Technology	Danna Freedman (Deputy Director)
Princeton University	Nathalie de Leon
Cornell University	Gregory Fuchs
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Fast and Cooperative Ion Transport in Polymer Based Materials (*FaCT*)
EFRC Director: Valentino R. Cooper
Lead Institution: Oak Ridge National Laboratory
Class: 2022 – 2026

***Mission Statement**: To understand and control fast, correlated ion and proton transport at multiple length and time scales in polymer-based electrolytes to overcome the current limitations of energy storage and conversion technologies.*

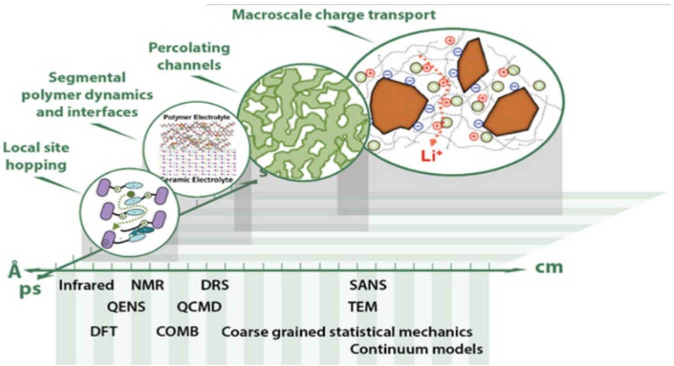

Figure 1 Length and time scales of experiments and theory relevant to the FaCT mission. (QENS = quasielasitc neutron scattering, BDS = broadband dielectric NMR = nuclear magnetic resonance, DFT = density functional theory, DRS = diffuse reflectance spectroscopy, QCMD = quartz crystal microbalance with dissipation monitoring, COMB = charge-optimized many-body potential, SANS = small-angle neutron scattering, and TEM = transmission electron microscopy.)

A major bottleneck limiting the advancement of energy storage and conversion technologies is the development of multifunctional, selective, and highly conductive membranes and solid electrolytes. Polymer-based electrolytes are amongst the most promising materials for next-generation applications due to their high electrochemical stability, mechanical flexibility, processibility, and their ability to maintain electrical contact over large temperature variations (good adhesion properties) better than their more brittle ceramic counterparts. Furthermore, they provide an ideal platform to support next generation energy storage architectures, such as those relying on the interdigitation of an electrolyte with the electrode. A grand challenge lies in the fact that polymer membranes, however, have yet to achieve the necessary conductivity and selectivity for fast ion transport and suffer from water management issues that restrict operating temperatures due to the need for water (i.e., hydrogen bonding networks) to transport protons. Efforts to improve conductivity, mechanical modulus, flexibility, and safety, by combining polymers and superionic ceramics, have thus far failed to provide the necessary conductivity.

This center seeks to build a predictive, data-driven, physics-based mechanistic model of ion and proton transport in polymers and polymer-ceramic composites to enable the targeted design of next-generation energy storage and conversion materials. *Materials-specific experimental and computational data covering a broad range of length and time scales will inform macroscale descriptions of charge transport to accelerate the design of polymer electrolytes with fast, correlated ion transport.* A foundational goal is to understand the nanoscale origins of ion and proton conductivity by identifying the mechanisms that determine how ions and protons move in polymers and composites on a microscopic (0.1–10 nm) scale and to correlate these movements to macroscale transport properties. In essence, we seek to discover new systems with superior charge transport to what is available today. Emphasis will be placed on determining (i) how ion diffusion/hopping can be decoupled from polymer segmental dynamics and how to significantly reduce the energy barriers for charge (alkali ions and protons) hopping in polymers; (ii) how conductivity is enhanced by specific ion channel morphologies and polymer solvation microenvironments; (iii) how ions move along or across dissimilar interfaces; and (iv) how specific chemical additives can accelerate charge transport and how they can be introduced in microstructured channels and at interfaces to improve conductivity. This will enable us to (i) increase ion mobility, (ii) improve ion solvation and dissociation, (iii) devise new chemical systems for fast hopping transport, and

(iv) promote fast, correlated charge transport in a material across nanometer to millimeter length scales. This research will concentrate on (i) small classical cations (Li^+ and Na^+) diffusing through a matrix, and (ii) protons where hopping transport and dynamic hydrogen bonding can be leveraged. As model polymer systems, we will initially focus on single ion conducting polymers and polymer-composites relevant to future clean-energy technologies. Single ion conductors provide selective ion transport and have significant advantage for many applications, in comparison with dual ion conductors. These systems are chosen to allow for one-to-one, seamless, comparisons, across multiple length and time scales, between theory and experimental synthesis and characterization efforts. To achieve our goal, *FaCT* is built upon two thrusts and a crosscut:

Thrust 1: Unveil the mechanisms driving correlated alkali ion and proton transport in polymers. The goal of this Thrust is to understand and realize new mechanisms of superionic conductivity (Li^+, Na^+, H^+) in polymer-based materials (i.e., > 1 mS/cm at room temperature). ***Key questions:*** What are the fundamental physics controlling charge transport in polymers and what are the microscopic mechanisms that determine energy barriers for ion hopping? Why does correlated ion motion suppress charge transport in polymeric materials, while it enhances charge transport in superionic glasses?

Thrust 2: Elucidate the mechanisms controlling correlated ion transport at the polymer–ceramic interface and reducing interfacial ion transport barriers. This Thrust seeks to elucidate the microscopic mechanisms controlling ion transport along and across polymer–ceramic interfaces, to design modified interfaces with reduced ion diffusion barriers. ***Key questions:*** How do interfacial chemistries and structures alter ion transport and dissociation? How do changes in interfacial polymer dynamics correlate with ion transport? What is the origin of the interfacial charge–transfer barrier and how can it be minimized?

Crosscut: Design through computation, theory, and materials informatics. The crosscut goal is to leverage fundamental knowledge obtained from the Thrusts to enable the design of fast ion conducting polymer and polymer-ceramic electrolytes. ***Key questions***: How can representation learning be utilized to combine experimental and simulation data to discover high-performing polymer electrolytes with superior charge transport? How can theoretical and computational approaches with vastly different data types be combined in an interconnected, seamless, multiscale manner to predict bulk and interfacial properties critical for fast, cooperative ion-conducting polymer-based electrolytes?

Fast and Cooperative Ion Transport in Polymer Based Materials (*FaCT*)	
Oak Ridge National Laboratory	Valentino R. Cooper (Director), Chelsea (Xi) Chen (Thrust 2 co-lead), Miaofang Chi (Crosscut co-lead), Rajeev Kumar (Thrust 1 co-lead), Alexei Sokolov (Thrust 2 co-lead), Sheng Dai, Tomonori Saito, Thomas (Zac) Ward
The Pennsylvania State University	Susan Sinnot (Deputy Directory), Michael Hickner (Thrust 1 co-lead), Wesley Reinhart (Crosscut co-lead), Ralph Colby
Georgia State University	Gangli Wang
Texas A. and M. University	Jodie Lutkenhaus (Thrust 1 co-lead)
University of California, Santa Barbara	Raphaële Clément
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https://www.ornl.gov/project/fast-and-cooperative-ion-transport-polymer-based-electrolytes-fact

Center for Molecular Electrocatalysis (CME)
EFRC Director: R. Morris Bullock
Lead Institution: Pacific Northwest National Laboratory
Class: 2009 – 2024

Mission Statement*: To establish cross-cutting principles that facilitate catalyst design for interconversion of electrical and chemical energy, across molecular and heterogeneous catalysis.*

CME will reveal the key mechanistic principles that unite and differentiate molecular and heterogeneous catalysis by providing an integrated thermodynamic framework broadly underpinning catalyst design. Through synergistic parallel efforts, we investigate molecular constructs both in solution and incorporated into electrode interfaces, while also controlling the environment beyond the binding site. (Figure 1).

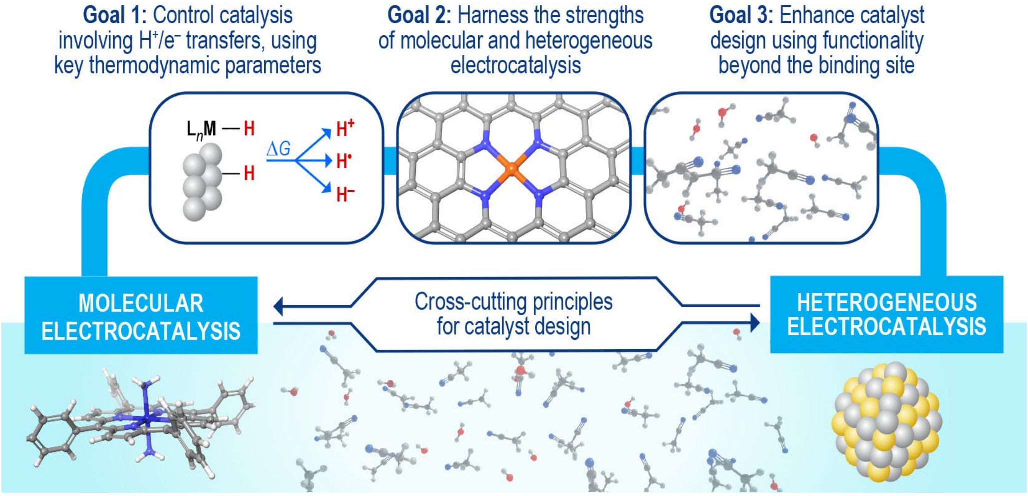

Figure 1. CME is developing cross-cutting design principles spanning molecular and heterogeneous electrocatalysis.

The impact of CME research will enable critical innovations in the design of catalysts. We seek to harness the most attractive features of molecular and heterogeneous catalysts through tailored coordination and solvation environments, such that highly active, efficient, selective, and durable catalysts can be designed for chemical transformations critical to energy conversions. As our research has evolved to include heterogeneous catalysis and interfacial studies, *our core principle has not changed: all catalysis is molecular, involving bond-breaking or bond-forming in molecules at active sites*, whether that site is in a soluble molecular complex, at a nanoparticle, or on a heterogeneous surface. Heterogeneous surfaces offer distinct attributes—including band-mediated charge transport and the ability to organize multiple cooperative active sites—that can be uniquely leveraged by the molecular perspective of CME. Our research is advancing catalyst design by unifying molecular and interfacial reactivity while mastering complex media, building upon our established strength in thermodynamic principles.

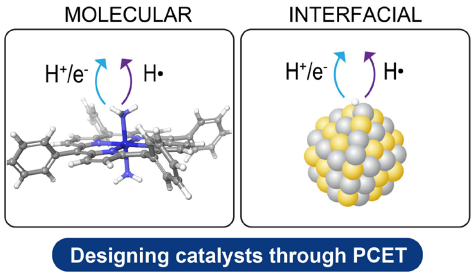

Figure 2. CME is using a proton-coupled electron transfer (PCET) framework as the basis for designing both molecular and interfacial catalysts through precise control of proton and electron transfers.

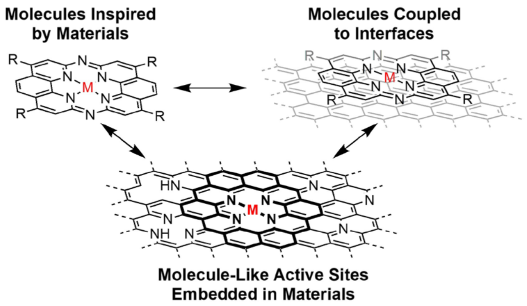

Figure 3. CME aims to bridge and combine principles from molecular and interfacial catalysis to develop new classes of molecules and materials for electrochemical energy

In the Thrust on **Designing Catalysts through a PCET Framework** (Figure 2), we are developing the fundamentals of energy-related electrocatalysis through a thermochemical PCET lens, combing experiment and theory. We are developing an understanding of the thermochemical landscapes of homogeneous and heterogeneous PCET and oxygen atom transfer processes and the implications of those landscapes for catalysis. Our Thrust on **Bridging Molecular and Interfacial Approaches to Electrocatalysis** seeks to develop unifying principles and predictive models that span and unite the traditionally disparate fields of molecular and heterogeneous electrocatalysis (Figure 3). Our research is exposing how interactions between a molecule and a polarized interface lead to distinct electrocatalytic reaction mechanisms and scaling behaviors. Our research Thrust on **Harnessing the Extended Catalyst Environment** develops the fundamental principles that underlie electrocatalytic processes at heterogeneous interfaces and with molecular catalysts in non-aqueous and low-aqueous media (Figure 4). We focus on how aspects of the catalyst system beyond the active site, including solvents, electrolytes, and redox mediators, affect the mechanisms and kinetics of electrocatalytic reactions to access new regimes of activity, selectivity, and stability.

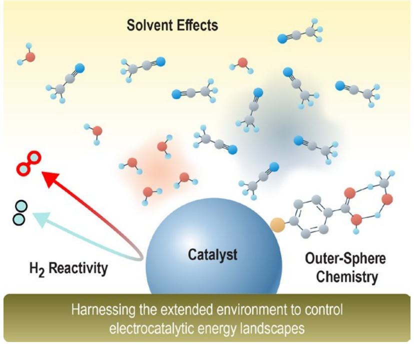

Figure 4. CME focuses beyond the binding site to develop design principles based on the extended environment, including the roles of solvents and outer-sphere interactions.

Center for Molecular Electrocatalysis (CME)	
Pacific Northwest National Laboratory	Morris Bullock (Director), Aaron Appel (Deputy Director for Operations), Simone Raugei (Lead, Theory/Computations), Eric Wiedner, Samantha Johnson
Yale University	Sharon Hammes-Schiffer (Deputy Director for Research), James Mayer (Thrust Lead, Designing Catalysts through a PCET Framework)
University of Wisconsin – Madison	Shannon Stahl, Thatcher Root
Massachusetts Institute of Technology (MIT)	Yogesh Surendranath (Thrust Lead, Bridging Molecular and Interfacial Approaches to Electrocatalysis)
University of Washington	Brandi Cossairt (Thrust Lead, Harnessing the Extended Catalyst Environment)
Caltech	Karthish Manthiram

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Interfacial Dynamics in Radioactive Environments and Materials (IDREAM)

EFRC Director: Carolyn I. Pearce
Lead Institution: Pacific Northwest National Laboratory
Class: 2016 – 2024

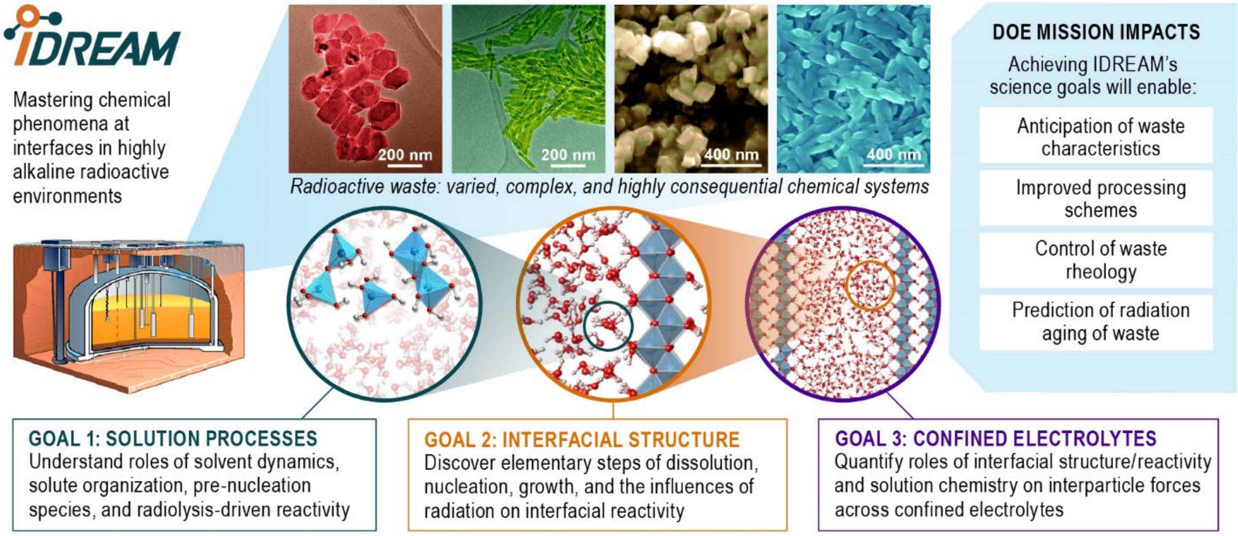
Mission Statement*: To master fundamental chemical phenomena and interfacial reactivity in complex environments characterized by extremes in alkalinity, low-water activity, and ionizing radiation.*

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 within the following Science Thrusts:

Science Thrust 1 (ST1) investigates Molecular and Solution Processes to understand solvent dynamics, solute organization, prenucleation species, and radiation-driven reactivity in concentrated alkaline electrolytes.
Science Thrust 2 (ST2) focuses on Interfacial Structure and Reactivity to discover the elementary steps of dissolution, nucleation, and growth and the influence of radiation on interfacial reactivity in highly alkaline systems of concentrated electrolytes.
Science Thrust 3 (ST3) examines the Dynamics of Confined Electrolytes to understand the chemical and radiation-driven phenomena of nanoscale-confined electrolytes that lead to interactions between interfaces to form aggregates of particles.

Our integrated crosscutting themes enable IDREAM discoveries across these three STs:

Crosscut 1 (XC1): Radiolysis and Radiation Dynamics. Fuses the knowledge emerging from our STs to facilitate a holistic understanding of the role of ionizing radiation in driving interfacial radiolysis in concentrated alkaline electrolytes.
Crosscut 2 (XC2): New Computational Tools and Theory. Provides an integrated theoretical basis to advance the study of hierarchical phenomena, reaction dynamics, and interfacial processes.
Crosscut 3 (XC3): Synthesis and Materials Characterization. Generates well-characterized materials and provides the foundation for their synthesis, including radiation-driven synthesis.



IDREAM focuses on the fundamental science that underpins the processing of millions of gallons of highly radioactive wastes stored at DOE's Hanford and Savannah River Sites.

IDREAM is the only EFRC focused on, and inspired by, the staggering chemical complexity of the legacy radioactive wastes that await processing. Weapons production at Hanford generated 56 million gallons of radioactive waste containing 170 million curies of radioactivity and 240,000 tons of complex chemicals. The waste is currently stored in huge underground steel tanks and is dominated by large quantities of aluminum in solution and in solid aluminum (oxy)hydroxide phases. Molar concentrations of sodium hydroxide, nitrate, and nitrite are also present, along with radioactive materials that provide a continuous supply of transient species. Waste processing is complicated by unknown "aging" mechanisms over time under extreme conditions of excessive alkalinity, concentrated electrolytes, and ionizing radiation. 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. Reducing the timeline to stabilize these wastes would have a considerable impact on the aggregate cost. IDREAM is enabling such cost reductions by advancing the study of complex electrolytes and aluminum (oxy)hydroxide suspensions and addressing the key knowledge gaps that limit waste processing. Examples of these fundamental studies include: (i) reducing uncertainty associated with precipitation kinetics of aluminum hydroxides; (ii) quantifying the effect of co-anions on aluminum solubility; and (iii) revealing the influence of solution composition on particle aggregation and settling behavior.

IDREAM integrates experiments, analytical probes, and computational modeling that span atomic to macroscopic length scales of solution speciation, interfacial reactivity, and confined electrolytes. Our efforts now also span a wide range of temporal scales, ranging from sub-picoseconds to weeks and months. Our synergistic research program has produced new fundamental knowledge about the chemical, physical, and radiolytic processes that occur in complex highly alkaline solutions and at interfaces. IDREAM's primary mission impact is Environmental Management and the enormous technological challenge of radioactive waste disposal. However, the impact of our scientific advances and novel capabilities extend to mission-critical challenges for DOE Basic Energy Sciences. The formation of complex mixtures of solution species and precipitating nanoparticles that aggregate underlies colloidal synthesis of nanostructured materials. Chemical reactivity at interfaces to form new ionized solution species or alter the solid is a feature of heterogeneous catalysis and electrochemical energy storage. The ability to predict solution speciation and manipulate it using solid substrates or external stimuli addresses challenges in critical materials recovery and CO_2 capture.

Interfacial Dynamics in Radioactive Environments and Materials (IDREAM)	
Pacific Northwest National Laboratory	Carolyn Pearce, Director Greg Schenter, Deputy Director Kevin Rosso, Zheming Wang, Xin Zhang, Jaehun Chun, Greg Kimmel, James De Yoreo, Michael Spradling
Utah State University	Aurora Clark
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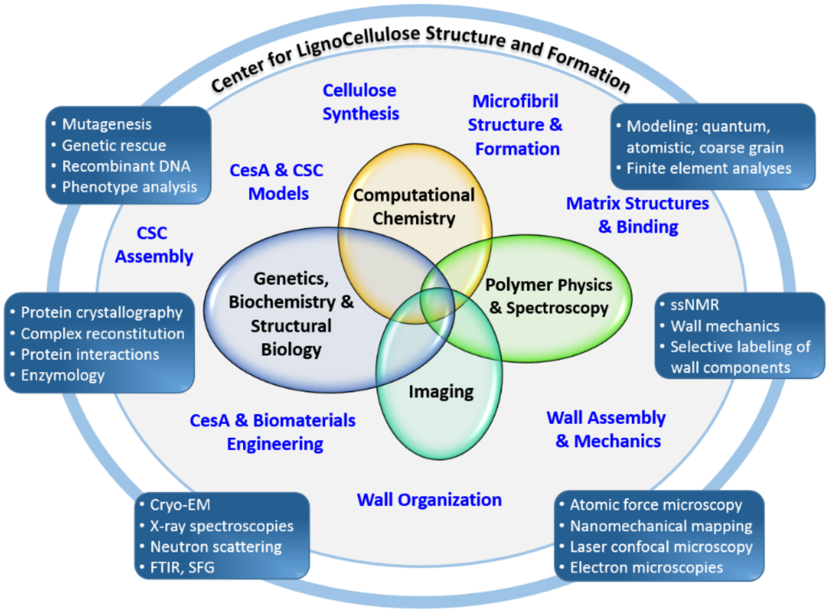
Center for Lignocellulose Structure and Formation (CLSF)

EFRC Director: Daniel J. Cosgrove
Lead Institution: The Pennsylvania State University
Class: 2009 – 2024

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).

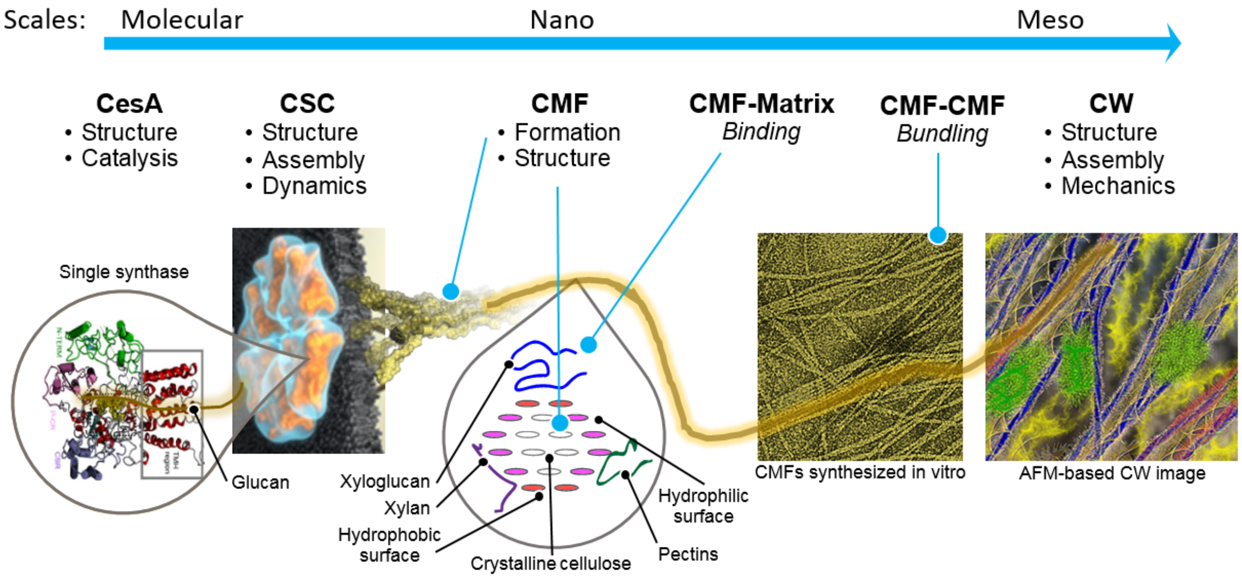
2. Manipulate active CesA assemblies *in vitro* and *in vivo* to learn how artificial and native CSCs are assembled and how cellulose microfibril structure depends on CSC structure. We will use these new experimental platforms to test computational models of CSC and cellulose microfibril assembly.

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.

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)	
The Pennsylvania State University	Daniel J. Cosgrove (Director), Charles T. Anderson, Enrique D. Gomez, Esther W. Gomez, Ying Gu, Seong H. Kim, B. Tracy Nixon, Amir Sheikhi
Massachusetts Institute of Technology	Mei Hong
North Carolina State University	Candace H. Haigler, Yaroslava G. Yingling
Oak Ridge National Laboratory	Hugh O'Neill
University of Cambridge	Paul Dupree
University of Rhode Island	Alison W. Roberts
The University of Texas at El Paso	James D. Kubicki
University of Virginia	Jochen Zimmer

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Center for 3D Ferroelectric Microelectronics (3DFeM)

EFRC Director: Susan Trolier-McKinstry
Lead Institution: Penn State
Class: 2020 – 2024

Mission Statement*: To develop three-dimensional, low-power, non-volatile ferroelectric memories that can be integrated reliably and densely interconnected with logic to enable low-power, 3D non-von Neumann circuits and systems*

The Center for Three-Dimensional Ferroelectric Microelectronics (3DFeM) is addressing fundamental scientific challenges required to enable beyond von-Neumann computing by densely interconnecting non-volatile memory with logic processors. The resulting step change in the connectivity of memory and logic will significantly reduce both the latency and energy cost associated with computation. To accomplish this, 3DFeM is probing the underlying mechanisms for emerging ferroelectric responses and tying the characteristic properties to the energy cost of computation, thereby addressing a significant US need by pursuing integrable, low-power on-chip memory. Ferroelectricity is attractive for low-power memories, as it allows access times <10 ns, retention exceeding 10 years, low power writing, high noise margins, and high endurance. While ferroelectric random-access memories have been commercialized by companies such as Texas Instruments, Cypress, Rohm, Panasonic, and Fujitsu, the available materials are limited in either thickness or coercive field scaling; this limits the scalability and density of memories that can be achieved. Moreover, the high processing temperature is an additional barrier for integration at the back-end-of-the-line (BEOL) in a semiconductor process, hindering 3D integration. However, recent developments in new ferroelectric materials point to the potential for a paradigm change. Exploring these defines the mission, goals, and research strategy of the center.

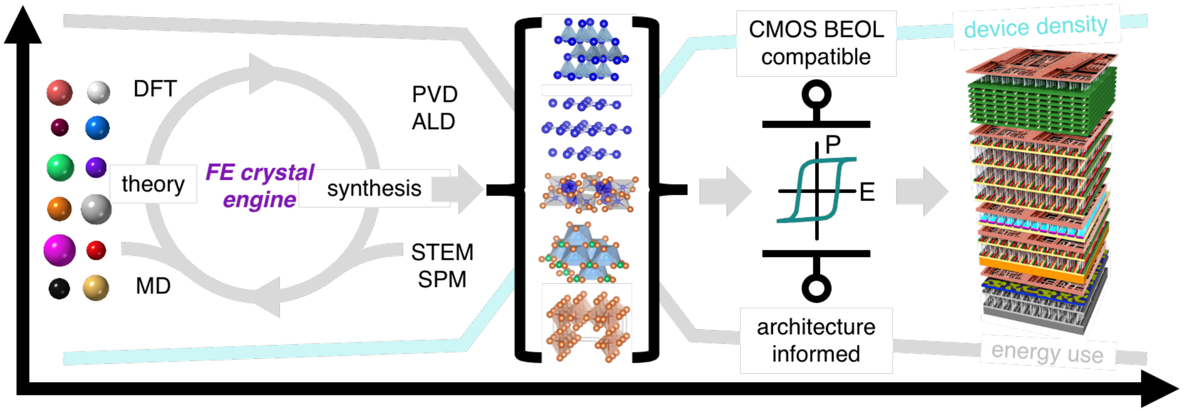

Fig. 1: The 3DFeM vision leverages fundamental understanding of emergent ferroelectricity created via nanoscale inhomogeneity to create memory devices densely interconnected with logic to enable low-power, 3D non-von Neumann computation

GOALS: 3DFeM will: (i) design ferroelectricity in new host crystal structures, (ii) tailor coercive voltages through engineering emergent nanoscale inhomogeneity in scaled ultra-thin films, (iii) deposit ferroelectric materials with ancillary electronics at low temperatures at wafer scale, (iv) characterize materials at previously inaccessible time and length scales, and (v) demonstrate device functionality.

RESEARCH STRATEGIES: 3DFeM is pursuing development of new ferroelectric materials that can be prepared at temperatures compatible with back-end-of-the-line processing, with tailored coercive fields, and robust reliability in memory applications. The goal, as shown in Figure 1 is to enable a significant increase in the connectivity between memory and logic that enables a significant reduction in energy usage. Computing accounts for 5 – 15% of worldwide energy consumption. While efficiency gains in hardware have partially mitigated the rising energy cost of computing, major gains are achievable in a paradigm shift to 3D computing systems. *3DFeM will reduce the energy cost of data movement while simultaneously manipulating data closer to where it is stored. This is now a critical need.*

3DFEM has built a synergistic team aligned towards accomplishing the center goals. The research in the center is organized around 2 Thrusts: *Designer Ferroelectrics for 3D Microelectronics* and *Integration and Reliability of 3D Ferroelectrics*. These two thrusts are closely intertwined, so that new understanding of basic mechanisms underpinning ferroelectricity and switching pathways can be developed, while simultaneously validating device functionality. Center-wide activities have developed common terminology, established healthy team dynamics, and prioritized tasks to accelerate research.

Thrust 1 serves as the designer ferroelectrics engine enabling 3D-integration of ferroelectric memory with processors. First, Thrust 1 seeks to identify "rules" for designing ferroelectricity by exploring the role of synthesis in stabilizing ferroelectricity in host structures normally considered to be only polar or centric. Second, the mechanisms responsible for polarization reversal in these new materials, including the switching pathways, will be identified. These efforts will culminate in the third goal of establishing new structure-property relationships governing ferroelectricity.

Thrust 2 integrates new ferroelectric materials into devices to assess their functionality and reliability as memory elements. For $Hf_{1-x}Zr_xO_2$ (HZO) and similar ferroelectric films, 3DFeM is enhancing our community's understanding of ferroelectricity and exploring opportunities for 3D integration. Nanoscale piezoresponse force microscopy (PFM) measurements have quantitatively demonstrated the role of oxygen stoichiometry on the phase assemblage, and hence the ferroelectric phase fraction and the wake-up characteristics for the first time. The collective domain dynamics also produce collective device responses. The proposed hardware-algorithm co-design utilizing the FeFET dynamics requires 1/3 fewer training samples (and hence 1/3 less energy) to converge compared to standard STDP.

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	Kyle Kelly, Nina Balke-Wissinger, and Stephen Jesse
Sandia National Laboratories	Thomas Beechem, Giovanni Esteves, Michael David Henry, Sean Smith
University of Tennessee	Sergei Kalinin

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Bio-inspired Light-Escalated Chemistry (BioLEC)
EFRC Director: Gregory Scholes
Lead Institution: Princeton
Class: 2018 – 2026

Mission Statement*: To combine light harvesting and solar photochemistry to enable more powerful editing, building, and transforming of abundant materials to produce energy-rich feedstock chemicals.*

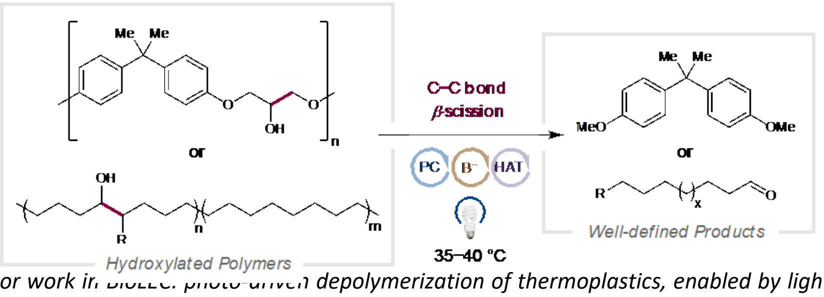
The search for more efficient, cost-effective, and green manufacturing technologies is of paramount importance. Chemical manufacturing accounts for 10% of total global energy consumption and is responsible for 7% of the world's greenhouse gas emissions. Fossil fuels remain the most widely employed source of energy to generate the high temperatures and pressures required to manufacture chemicals, and are often also used as starting materials for production. Solar energy can be converted to chemical energy when used to excite light-absorbing catalysts in chemical syntheses in a manner akin to that of photosynthesis, and transformation of abundant biomass to feedstock chemicals could alleviate the need for non-renewable sources; BioLEC aims to tackle both these challenges.

In contrast to traditional synthetic routes, photoredox catalysts utilize light to drive reactivity under very mild conditions. These methodologies have been mostly employed in the synthesis of small molecules such as pharmaceuticals, and are yet to be translated into mass production of feedstock chemicals. Photocatalytic methods, though highly efficient and mild, proceed via intricate mechanisms that are not easily applicable in broad scope large scale reactions. Translating these technologies to a wider range of chemical manufacturing applications therefore requires new approaches that make the reaction steps more robust and more efficient, as well as the underlying mechanistic knowledge to target these steps.

Since launching in August 2018, BioLEC has made significant advances on multiple fronts: in understanding what drives photoredox catalysis at a mechanistic level to enable the design of new improved catalysts; in taking inspiration from photosynthesis to tackle kinetic, efficiency and specificity shortcomings in traditional photocatalysts, such as obviating the low absorption cross-section limitation of organometallic photocatalysts using energy transfer from light-harvesting systems; in using light to manipulate enzymes for innovative photochemistry; and in employing photocatalysts for novel reactions for energy applications, such as light-driven recycling of polymers to reusable monomer building blocks, and the contrathermodynamic photocatalytic isomerization of internal olefins to more reactive terminal olefins. As we enter this second phase of our center, we will advance towards our goals through three thrusts:

A: Develop innovative photochemistry that enables new routes for synthesizing chemical feedstocks

We will develop new photochemistry and feedstocks in BioLEC, and expand the scope of mechanistic tools and reactive substrates available to the photocatalysis community. Drawing on the rich physical chemistry capabilities in BioLEC, new photochemical techniques for reactivity and

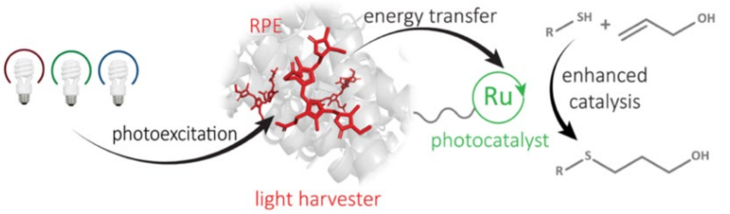


Prior work in BioLEC: photo-driven depolymerization of thermoplastics, enabled by light-driven O–H proton-coupled electron transfer. Approach is compatible with insoluble polymers and involves no stoichiometric reagents.

experimental characterization will be developed. New reactivities will push the boundaries of photoexcitation and accessible redox potentials. Finally, this new chemistry will valorize feedstocks that were previously inaccessible to photochemistry, providing a pathway toward recycling waste.

B: Discover, synthesize, and study photoenzymes that enable enhanced catalysis

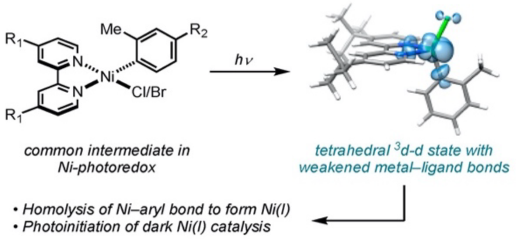
BioLEC's work transforming enzymes' catalytic abilities with light provides not only innovative functionality but also key insights into the fundamental functions of the microenvironment within the protein. By designing microenvironments in photoactive proteins, or "photoenzymes", we will introduce functionality that first mimics and then exceeds that of natural systems. Additionally, bioinspired tactics from photocatalysis involve separating catalytic functions to improve efficiency or selectivity.



Prior work in BioLEC: Light-harvesting protein conjugated to Ru photocatalyst generates increased yields and makes reactions at previously unusable irradiation wavelengths possible.

C: Inform design of photocatalysts by elucidating photocatalysis mechanisms

In the first phase of BioLEC we advanced fundamental knowledge of the elementary organometallic steps available through visible light excitation by applying a variety of physical techniques (e.g., transient absorption spectroscopy, organometallic synthesis, EPR, pulse radiolysis, ultra-fast IR) to study the mechanisms in multiple metallaphotoredox reactions, including C–O, C–N, and C–C couplings. We will build on these efforts with the goal of gaining a more global understanding of how changes to substrate, ligand, and photocatalyst identity affect the reaction mechanism. We will focus on using this understanding to design improved and new dual transition metal/photoredox coupling reactions.



Prior work in BioLEC: Ni(II) excited states relevant to Ni-photoredox catalysis

Bioinspired Light-Escalated Chemistry (BioLEC)	
Princeton University	Gregory Scholes (Director), Paul Chirik, Robert Knowles, David MacMillan, Barry Rand
Massachusetts Institute of Technology	Gabriela Schlau-Cohen (Associate Director)
Arizona State University	Ana Moore, Thomas Moore
Brookhaven National Laboratory	Matthew Bird
Cornell University	Todd Hyster
Michigan State University	James McCusker
National Renewable Energy Laboratory	Garry Rumbles
North Carolina State University	Felix Castellano, Elena Jakubikova
Northeastern University	Sijia Dong, Hannah Sayre
SLAC National Accelerator Laboratory	Amy Cordones-Hahn, Kelly Gaffney
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Photonics at Thermodynamic Limits (PTL)
EFRC Director: Shanhui Fan
Lead Institution: Stanford University
Class: 2018 – 2024

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

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

Objectives for 2018-2022

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

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

Center Research Team and Scientific Organization

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

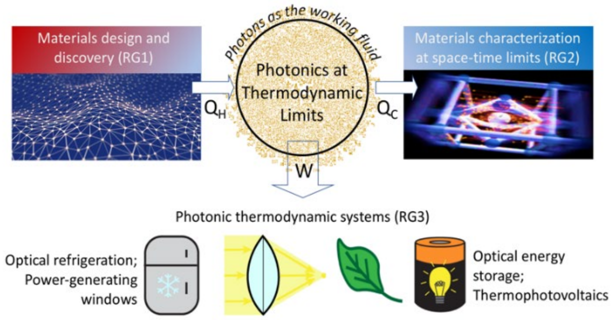

***Figure 1:** Schematic overview of our “Photonics at Thermodynamic Limits” Energy Frontier Research Center.*

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

- **Research Group 2 – Materials Characterization at Space-Time Limits:** The RG2 team is developing transformative characterization methods that include (1) novel in-operando characterization platforms based on optical microscopy within a transmission electron microscope (OTEM); (2) ultrafast stimulated emission depletion (STED) optical microscopy; (3) photothermal deflection spectroscopy; and (4) single particle ultrafast diffraction leveraging the unique facilities at SLAC National Accelerator Laboratory.
- **Research Group 3 – Photonic Thermodynamic Systems:** The RG3 team will investigate emergent physical phenomena and photonic 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.

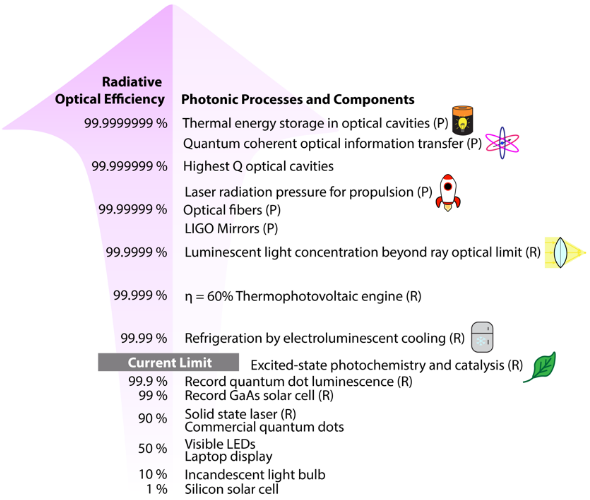

Figure 2: *Passive (P) and Active radiative (R) systems that become possible with increasingly high optical efficiency.*

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)	
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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 Unconventional Formations (CMC-UF)
EFRC Director: Anthony R. Kovscek
Lead Institution: Stanford University
Class: 2018 – 2024

Mission Statement*: to garner cross-cutting geoscience knowledge to achieve mechanistic control over the strongly coupled nonequilibrium physical and geochemical processes in extreme geological environments including shale, mudstone, marls, and other tight rocks with nanoscale pores. Collectively, these are referred to as unconventional formations and all subsurface storage formations are sealed by an unconventional formation.*

Unconventional formations will play an outsized role in the transition of energy from large greenhouse gas emissions to renewables and net-zero emission technologies in the coming decades. Importantly, unconventional formations serve as the geological seals atop conventional subsurface formations, Fig. 1-1, in addition to being a significant source of low-carbon intensity natural gas. This functionality implies confinement of sequestered CO_2 and storage of intermittent renewable energy (e.g., green hydrogen or compressed air) beneath seals as well as isolation of nuclear waste. While unconventional formations are ubiquitous throughout the subsurface, our fundamental and engineering science knowledge of them is limited by their nanoporous structure, extreme heterogeneity that spans at least 10 orders of magnitude spatially, and significant reactivity. Four challenges inhibit a system-level understanding of unconventional formations: (1) the physics of fluid transport and phase behavior are poorly understood due to dominant molecular interactions between rock/fluid molecules under confinement; (2) their mechanics, especially in the presence of nonaqueous fluids such as H_2 and CO_2, requires significant exploration; (3) reactivity of solid/fluid interfaces is governed by complex processes that are strongly coupled to transport and mechanics; and (4) the lack of scale separation prevents a reliable (closed) description of physicochemical processes at any single scale of observation. Our fundamental science goals G1—G5, and research plan below, address these challenges directly.

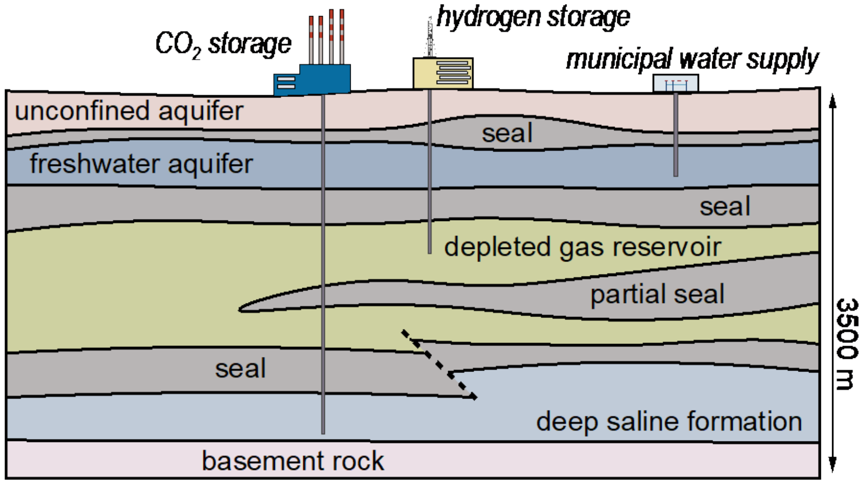

Fig. 1-1. Typical geological strata with seals (unconventional formations). Gas reservoir is repurposed for seasonal H_2 storage. The reservoir seal prevents vertical H_2 movement. Anthropogenic CO_2 is injected into a deep saline formation for secure storage. Top and bottom seals confine CO_2 to the formation.

Seal integrity and storage capacity are subject to geochemical interactions at shale-mineral interfaces, and confined phase and transport behavior that emerge out of intricate interactions in the mechanically stressed nanoporous structure. This structure is heterogeneous across scales, from nm to m. The fundamental understanding that is needed to assemble this complex puzzle requires exploration of the impact of microscale heterogeneity and exploiting transformative advances in experimental and computational modeling capabilities across scales. These are important to unraveling the role of larger-scale structural heterogeneity and variability that could be important for real-world full-scale projects including faults and weaker sections of a cap rock. CMC-UF's **approach is multiscale and multiphysics** while incorporating and integrating experiments, characterization before and after reactions, theory, and computational modeling efforts. Our research expertise is interdisciplinary, collaborative, and aligned with our science goals (**G1—G5**): (1) multiscale characterization and imaging; (2) multiphase, multiphysics transport processes; (3) reactivity at fluid/solid interfaces; (4) geomechanics; and (5) scale translation.

CMC - UF	
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University of Wyoming	Vladimir Alvarado, Saman Aryana, Teresa Lehmann
University of Southern California	Kristian Jessen, Theo T. Tsotsis
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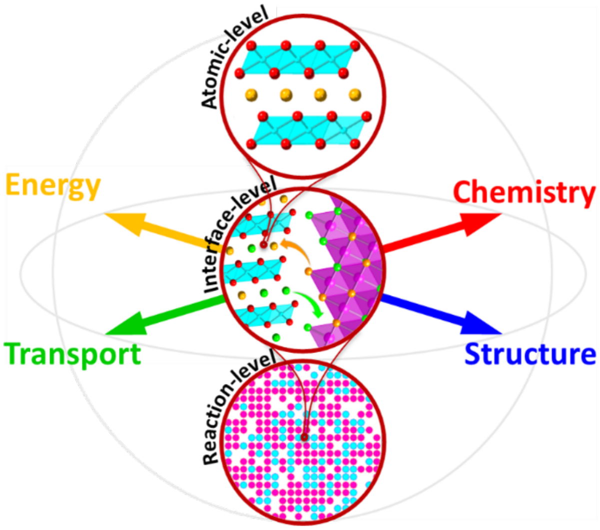
A Next Generation Synthesis Center (GENESIS)
EFRC Director: John B. Parise
Lead Institution: Stony Brook University
Class: 2018 – 2024

Mission Statement*: "To elucidates how key structural and chemical transformations are governed by the flow of energy and atoms across multiple length scales by integrating advanced in situ diagnostics and data science tools to interrogate and predict the pathways that govern synthesis and lead to new materials.*

We will further develop and integrate the computational and *in situ* experimental infrastructure to address precision synthesis of bulk materials systems. For polycrystalline inorganic solids, our aim is to produce materials that have homogeneous local and long-range crystalline structures. At the *atomic scale*, this requires control of atomic configurations that are desired, along with control of composition, structural defects and impurities. At the *mesoscale* (interfacial, particle- and reaction-scales), precision synthesis involves the ability to control chemical microenvironments, relative kinetics, heterogeneity, compositional gradients, and microstructure.

We will study model systems to resolve synthesis pathways and interrogate how the composition, structure, and heterogeneity of multi-component reactive system evolve through a thermodynamic landscape. Developments of *in situ* diagnostics that span Å-to-mm length scales will be leveraged to build a fundamental understanding of the interplay between thermodynamic driving forces, reaction pathways and kinetics, and transport kinetics, which are all affected by the synthesis conditions as well as the choice of precursors and the properties of resulting product. The understanding gained, through both computational and experimental investigations, will ultimately enable *de novo* prediction and adaptive control of synthesis routes.



In GENESIS, we aim to unravel this complexity by emphasizing understanding and control on the atomic-scale and mesoscale phenomena, as well as how they impact the reaction outcome. In the picture of synthesis as the flow of atoms and energy from reagents to products, we focus on making and breaking bonds, along with the redistribution of atoms and energy between these. GENESIS see the need to develop new tools to drive new understanding in synthesis and will continue these efforts, where next generation computational and experimental tools will be developed, benchmarked, and integrated with the previous successful developments and current ongoing efforts.

For Atomic-Level Control. The preparation of many bulk inorganic solids is carried out by either reacting mixtures of precursor powders at high temperature or through precipitation from solution. High temperatures facilitate diffusion in the solid state and drives reactions towards a thermodynamic minimum. However, a wider range of metastable material products exist at local minima in the reaction landscape. Accessing these metastable products (intermediates with respect to the global minimum) requires kinetic control, achieved for example, through defect engineering, precursors design, or solution routes. This thrust will focus on kinetically controlled reactions in metastable phases and phases incorporating metastable defects within a thermodynamically stable product. The key questions being addresses in studies of atomic scale phenomena: *What defines the point of differentiation between forming structure A vs structure B? How does this "tipping point" differ for different pathways leading to the same product? What is the relative importance of precursor vs conditions? What governs the range of different compositions that can be achieved for the same structure? Do reactions from isomorphic precursors to isomorphic products follow parallel pathways?*

For Mesoscale Control. The synthesis of solid inorganic materials is intrinsically non-uniform, involving heterogeneous mixtures of particles of different material phases. Reagents with one set of structures and compositional distributions evolve to form products with a different set of structures and compositional distributions. This evolution involves correlated parameters, across multiple length scales, from atoms to interfaces to reactive particle aggregates. Progress will require new tools capable of interrogating systems at multiple length scales to 1) evaluate native morphological features and how these evolve, and 2) manipulate morphology and evaluate difference in reaction evolution. These capabilities are critical to controlling syntheses and ultimately for the eventual scale-up of bulk materials. The key questions: *How does a reaction evolve from an interface and between particles? What heterogeneity exists and on what scale, and can it be controlled through, eg. reagent architecture? Can we control transport and reaction outcomes by designing pairwise reactions or with tailored architecture? Do preferential transport directions and lowest surface energies impact outcomes? Is the reaction enhanced at the higher energy faces? What are the effects on nucleation? Can we harness surface effects such as mobility and melting?*

Advancing Tools. GENESIS will develop and integrate new tools across the spectrum of theory, simulation, data analytics, and experimentation to elucidate the role of defects, heterogeneity, and microstructure in synthesis to address the science questions above. We focus on tool development and their integration to provide understanding from data, and to develop active control of reactions. The new characterization tools will complement the suite of capabilities GENESIS has already developed and utilized. While we have begun to examine the effects of architecture in how reactions proceed, incorporating the role of defects into such models requires theory and atomistic calculations of energetics and kinetic properties of defects and amorphous materials.

A Next Generation Synthesis Center (GENESIS)	
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Colorado State University	James Neilson
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Center for Mesoscale Transport Properties (m2m#S)
EFRC Director: Amy Marschilok
Lead Institution: Stony Brook University
Class: 2014 – 2026

***Mission Statement**: To understand and harness disorder and entropy to build the science foundation for new design spaces that enable sustainable, long cycle life electrochemical energy storage.*

Project Objectives

Batteries are enabling technologies for both transportation and large-scale storage as the energy landscape is electrified. To meet the needs, new battery systems must be imagined, designed, and implemented. However, a universal bottleneck for nearly all "post-Li-ion" batteries is unsatisfactory cell cycle life. This renewal proposal will build the scientific foundation of a broadened design space to enable future creation of long-life batteries based on earth abundant materials. The advancement requires understanding and controlling both kinetic and thermodynamic properties. In the renewal, the Center for Mesoscale Transport Properties (m2m#S) mission will be realized by exploiting entropy and disorder, exploring electrochemical mechanisms beyond lithium ion insertion, and designing and controlling resilient interfaces.

Science Goals

The Center will integrate synthesis, characterization, theory, modeling, and electrochemistry to achieve 3 science goals (**Figure 1**).

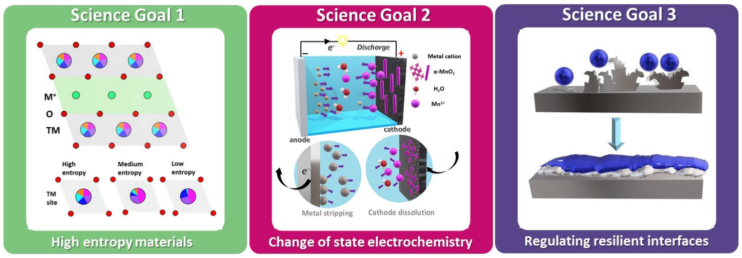

Figure 1. *m2m#S* Center Science Goals (SG).

1. Exploit entropy and disorder as guiding principles toward understanding of new earth abundant electroactive materials.
2. Understand and control mechanisms involving ordered ⇋ disordered changes of state (deposition/dissolution, plating/stripping) as active electrochemical processes in sustainable systems.
3. Control interfacial order through design and fabrication of resilient interfaces.

Summary of Proposed Research

New battery systems are critically needed to serve the expanding needs for energy storage. Thus, novel approaches to materials and electrochemistry must be imagined, designed, and implemented, making more elements of the periodic table accessible for productive electrochemical energy storage. A recent class of multicationic materials classified as high entropy oxides (HEOs) has emerged analogous to high entropy alloys. The phase stability of the multicomponent system can be estimated using the Gibbs–Helmholtz equation: $\Delta G\text{mix} = \Delta H\text{mix} - T\Delta S\text{mix}$, where $\Delta G\text{mix}$, $\Delta H\text{mix}$, and $\Delta S\text{mix}$ are the changes of the Gibbs free energy, mixing enthalpy, and mixing entropy, respectively, and T is temperature. A single-phase crystal structure benefits from entropy stabilization by increasing the configurational entropy through including multiple elements, randomly distributed on the same lattice sites. High entropy materials often exhibit resilience, including high fracture toughness, high strength, good high/low temperature function, and energy storage properties. There is strong motivation to find a suitable approach for the synthesis of pure materials with tunable composition, size, and morphology to provide a rich design space for new electrochemically active substances.

A universal bottleneck for nearly all so-called “post-Li-ion” batteries is the unsatisfactory reversibility of their phase transitions leading to limited cell lifetime. All batteries of contemporary interest employ at least one, oftentimes two, solid-state electrodes. This choice is in part due to the higher volumetric energy density and handling convenience. Thus, a common feature of nearly all solid-state electrode materials is their crystallinity that dictates the physical properties, reactivity, and directional responses of macroscopic materials, with only a few exceptions. A consequence is that the charge-discharge cycling of any rechargeable battery cell hinges upon the reversibility of the first-order, structural phase transitions of the solid-state materials of the battery electrodes. Thus, new electrochemical concepts beyond simple insertion of lithium ion will be pursued.

Key to the effective function of all batteries is formation of effective interphases that permit productive ion transfer while blocking parasitic decomposition processes. In active batteries, multiple substances are in contact where the solid-liquid and solid-solid interfaces (among electroactive materials, electrolytes, conductive additives, and binders) may experience dynamic chemical/mechanical changes leading to compromised mesoscale stability. A frequently occurring issue is thermodynamic incompatibility upon (dis)charge resulting in interfacial chemical and electrochemical decomposition with accompanying irreversible chemical reactions leading to high local resistance or mechanical damage resulting in loss of electrical connection. New materials will be explored to provide sufficient mechanical strength, appropriate conductivity, and cost-effective scalable fabrication methods.

Scientific Impact of Proposed Research.

The ability to manipulate material composition and to use entropy (S) productively advances fundamental thermodynamic understanding and holds the potential for broad impact on many fields. Consideration of the factors dominating ion-electron coupled mechanisms beyond ion insertion is necessary to transition electrochemical energy storage into new paradigms. Finally, the ability to deliberately form and modify interfaces will impact any electrochemical processes often dominated by the nature of an electrode surface.

Center for Mesoscale Transport Properties (m2m#S)	
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Columbia University	Alan West
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Reconfigurable Electronic Materials Inspired by Nonlinear Neuron Dynamics (REMIND)
EFRC Director: R. Stanley Williams
Lead Institution: Texas A&M Engineering Experiment Station
Class: 2022 – 2026

Mission Statement*: To establish foundational scientific knowledge underpinning the function of massively reconfigurable computing architectures that approach fundamental limits of energy efficiency and speed, enabling real-time learning and embedded intelligence emulative of specific neuronal and synaptic functions of the human brain.*

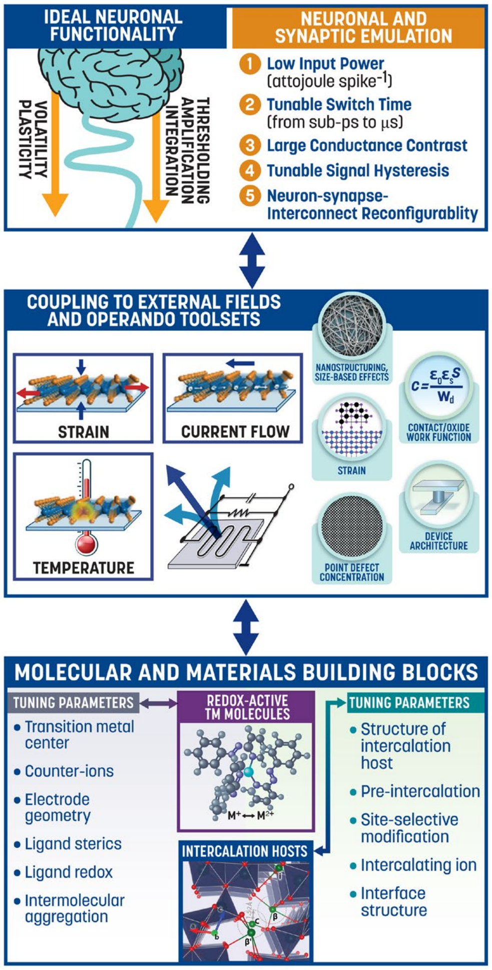

***Fig. 1. REMIND** will create materials for artificial neurons and synapses by controlling the physics and chemistry of nonlinear dynamical electronic transitions to provide a fundamental basis for neuromorphic computing.*

The current paradigm of developing computational devices and architectures invokes forward design, which leverages the well-defined properties of silicon and its interfaces with metals and dielectrics to tune electrical conductance *via* well-established mechanisms. **REMIND's** goal is to connect dynamical material properties and underlying electrochemical transformations to discover and exploit new mechanisms, materials, and interfaces that can emulate specific neuronal and synaptic functions of the human brain. We will design and realize reconfigurable neuron- and synapse-like materials based on crystalline transition metal oxide intercalation hosts and thin films of self-assembled redox-active coordination complexes with precisely programmable redox cascades. We will flip the current paradigm by blending inverse and forward design and connecting dynamical material properties and underlying transformations to discover and exploit new materials, mechanisms, and interfaces that are required to emulate specific neuronal and synaptic functions (**Fig. 1**).

REMIND comes at a watershed moment for computing. Transmitting, storing, and processing data already account for approximately 10% of global energy use. Fifty zettabytes of data were collected in 2020, and that very large number (a zettabyte is 10^{21} bytes) is expected to grow a million-fold by 2040. The microelectronics industry is encountering roadblocks directly traceable to fundamental physical constraints of present computing paradigms. This upward trajectory of data has just begun in earnest, Moore's Law scaling has stalled, and the efficiency of transistors has not improved in the past few generations, thus setting the stage for a crisis in computing and the global information economy.

REMIND's goals are to: (**1**) Identify fundamental neuromorphic conductance switching mechanisms that enable 4 to 5 orders of magnitude improvements in speed and energy efficiency of neuromorphic analog computing over scaled digital CMOS. (**2**) Develop experimental tools to interrogate form and function, bridging length, time, and energy scales, to predict the cumulative nonlinear electrically-triggered response of (a) microscopic elements and (b) nontrivial ensembles of such elements interfaced within physical networks. (**3**) Create inverse design rules that work backwards from neuronal/synaptic function to material/interfacial properties (**Fig. 1**). (**4**) Tailor conductance switching and reconfigurability across two material classes that exhibit low-entropy transformations: (a) intercalation hosts and (b) transition-metal coordination complexes, using materials design, host-guest chemistry, site-selective modification, and lattice strain (**Fig. 2**) to establish rules for decoupling transformation characteristics such as conductance differential, threshold voltage, sharpness of the transition, and hysteresis. (**5**) Demonstrate *in situ* device reconfiguration by tuning material properties, thereby enabling small-scale networks that can explore the ultimate limits of speed and energy consumption.

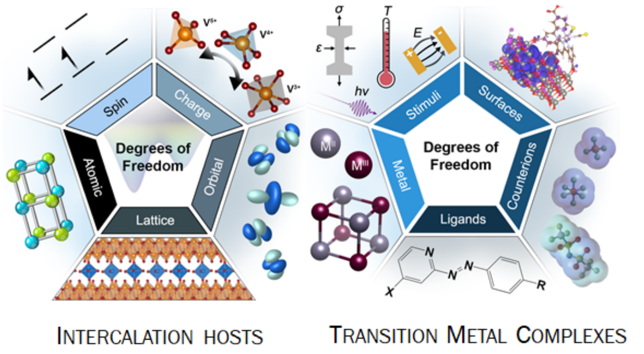

***Fig. 2. REMIND** will explore, expand, and navigate the design space of electronic instabilities using materials design, host-guest chemistry, site-selective modification, and lattice strain.*

REMIND's materials chemistry focus areas investigate low-entropy electrochemical transformations in intercalation hosts and thin films of redox-active molecular complexes as a means of neuronal and synaptic emulation (**Fig. 2**). Our multidisciplinary team will leverage DOE national user facilities to research new molecules, materials, interfaces, and circuit elements by integrating operando toolsets for interrogating the dynamics of electronic and atomistic structure, scale-bridging modeling that captures discontinuous changes in electronic properties, and machine learning and artificial intelligence frameworks mapping desired neuronal and synaptic function to the structure and composition of molecules and materials.

Success will be a new scientific foundation for neuron- and synapse-like materials that enable intelligent and energy-efficient information processing inspired by living systems. The knowledge gained will pave the way to computing architectures with unprecedented efficiency, speed, and reconfigurability; "reboot" the microelectronics industry; and through enablement of real-time AI, will lay the foundations for a new era of atom-precise transformative manufacturing.

Reconfigurable Electronic Materials Inspired by Nonlinear Neuron Dynamics (REMIND)	
Texas A&M Engineering Experiment Station	R. Stanley Williams (Director), Sarbajit Banerjee (Executive Associate Director), Perla Balbuena, Kim Dunbar, Marcetta Darensbourg, Raymundo Arroyave, Matt Pharr, Patrick Shamberger, Xiaofeng Qian
National Renewable Energy Laboratory	Andrew Ferguson (Associate Director), Jeffrey Blackburn, Lance Wheeler, Katherine Jungjohann
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Center for Soft PhotoElectroChemical Systems (SPECS)

EFRC Director: Erin Ratcliff
Lead Institution: University of Arizona
Class: 2022 – 2026

Mission Statement*: SPECS will understand the factors controlling charge and matter transport processes in inexpensive, scalable, and durable π-conjugated polymer (plastic) materials. We will explore the factors across spatiotemporal scales that underpin emerging energy conversion technologies to influence the formation of fuels, such as H_2, from sunlight and develop new approaches to energy storage.*

The diverse and engaged SPECS team includes synthetic, computational, and experimental experts working across three interconnected thrusts (**Figure 1**) involving π-conjugated, polymeric (plastic) semiconductor materials: **Hybrid Electrical-Ionic Charge Transport** (Thrust 1), **Charge Transfer and Energy Cascades** (Thrust 2), and **Durability** (Thrust 3). SPECS will fill crucial knowledge gaps that limit the introduction of these materials into the U.S. energy portfolio including: i) understanding and control of interface molecular composition and structures dictating charge (electrical and ionic) transport, ubiquitous across all electrochemical energy conversion and energy storage applications; ii) understanding dynamic responses of interfaces and mechanisms that drive (photo)-electrochemical charge transfer processes; and iii) creation of design guidelines that enable robust π-conjugated polymeric energy conversion and storage materials.

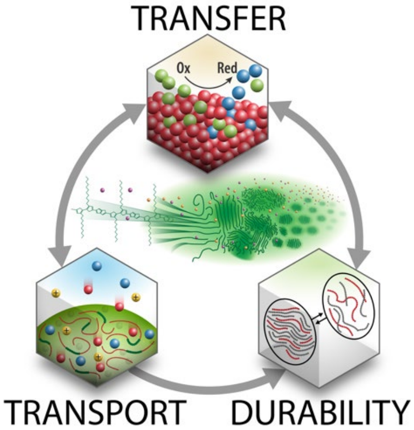

Figure 1. SPECS thrusts working synergistically to understand charge and matter transport processes in soft materials.

Direct outcomes of SPECS will be mechanistic insights and new physiochemical models of fundamental structure-property relationships of π-conjugated polymer platforms. Models based on fundamental chemistry will drive the search for anode and cathode materials at electrochemically reactive interfaces (catalysis) and buried atomic-to-nano interfaces that comprise the bulk (storage). These mechanistic insights require developing new measurement science approaches, which will be a high impact legacy of SPECS. We will advance characterization and operando approaches, under both environmental and operational "stresses," examining materials in near-surface, interface, and interphase regions, under conditions that push soft materials and platforms far from equilibrium. Fundamental insights gained by SPECS will translate across *Science for Clean Energy* applications, including connections to existing BES (EFRC and Hub) programs. SPECS research products will be disseminated to the greater scientific community through high-impact journal articles and creation of new intellectual property. SPECS represents a scientifically and demographically Diverse and Inclusive Team that fosters new knowledge discovery coupled to unique mentoring and student training, targeted to development of an inclusive next generation workforce needed to meet U.S. 2050 clean energy goals as well as Energy Justice initiatives.

To achieve SPECS' goals, we are organized into three synergistic and interconnected research thrusts. Thrust 1 focuses on comprehensive characterization of the complex π-conjugated polymer/electrolyte structures that control ion and polaron transport and energy storage processes. We will emphasize understanding fundamental design principles that drive the dynamic response (picosecond-to-second) of these systems at atomic-to-micron length scales. Thrust 1 includes a tight feedback loop among theory/modeling, materials design, synthesis, processing, and characterization to link complex electronic, electrochemical, and physical phenomena. Objectives include: 1.1) describe the physical structure of polymer-electrolyte interphases and how dynamic versus static structural disorder affects relevant transport processes, 1.2) define structure-property relationships controlling hybrid electrical-ionic transport and rates of charge/discharge in polymer-electrolyte systems, and 1.3) quantify strengths of ion-polaron interactions and describe how these interactions impact hybrid electrical-ionic transport.

Thrust 2 will understand and design π-conjugated polymer photocathodes that are tailored for efficient, directional electron and hole transfers as well as long-lived charge separation needed to drive charge (energy) for fuel-forming reactions, such as H_2 evolution. Data from theory/modelling, spectroscopy, and structural characterization will be critical to elucidate mechanisms of energy and charge transfer that we will leverage to benchmark and improve performance in relevant electrochemical π-conjugated polymer platforms. Within Thrust 2, SPECS aims to tailor interactions of complex phenomena to achieve integrated multicomponent systems. This thrust is tightly linked to efforts of Thrust 1 and ultimately Thrust 3 to ensure new durable energy conversion materials. Objectives include: 2.1) define the equivalent of an electric double layer in increasing complex π-conjugated polymer/electrolyte interfaces under operando (applied bias and/or light), 2.2) describe mechanisms of electron transfer in outer sphere redox reactions, and 2.3) investigate photoelectrochemical mechanisms — leading to solar fuel formation (H_2).

Thrust 3 will establish molecular scale understandings of π-conjugated polymer degradation and its mitigation, leading to electrochemical/photoelectrochemical systems that simultaneously exhibit high performance and long lifetimes. Durability of these materials is determined by a combination of bond forming/bond breaking, microstructure, and long-range connectivity of mechanical, thermal, optical, electronic, and electrochemical properties. Thrust 3 objectives include: 3.1) determine the chemical degradation pathways and eliminate detrimental chemistries, 3.2) evaluate long-time, operando-induced morphological changes and mechanical failures providing design guidelines to improve durability, and 3.3) manipulate local chemical and structural environments to increase durability. These three thrusts working cooperatively together will allow SPECS to gain a fundamental understanding of low-cost, durable energy conversion and energy storage technologies leveraging soft materials.

SPECS	
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National Renewable Energy Laboratory	Elisa Miller (Deputy Director), Garry Rumbles (Senior On-Site Advisor), Annie Greenaway, Andrew Ferguson,
University of Colorado, Boulder	Seth Marder (Durability Thrust Co-Lead), Mike Toney, Steve Barlow, Obadiah Reid
Emory University	Tianquan (Tim) Lian (Charge Transfer Thrust Lead and Associate Director of Scientific Continuity)
University of Kentucky	Chad Risko (Charge Transport Thrust Lead)
Georgia Institute of Technology	Natalie Stingelin (Associate Director of Scientific Continuity)
Purdue University	Jianguo Mei (Durability Thrust Co-Lead)
Stanford University	Alberto Salleo

Contact: Erin Ratcliff, SPECS Director, ratcliff@arizona.edu
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Manipulation of Atomic Ordering for Manufacturing Semiconductors (μ-ATOMS)

EFRC Director: Shui-Qing (Fisher) Yu
Lead Institution: University of Arkansas, Fayetteville
Class: 2022 – 2026

Mission Statement: *To discover the underlying science principles determining the ordering of atoms in semiconductor alloys.*

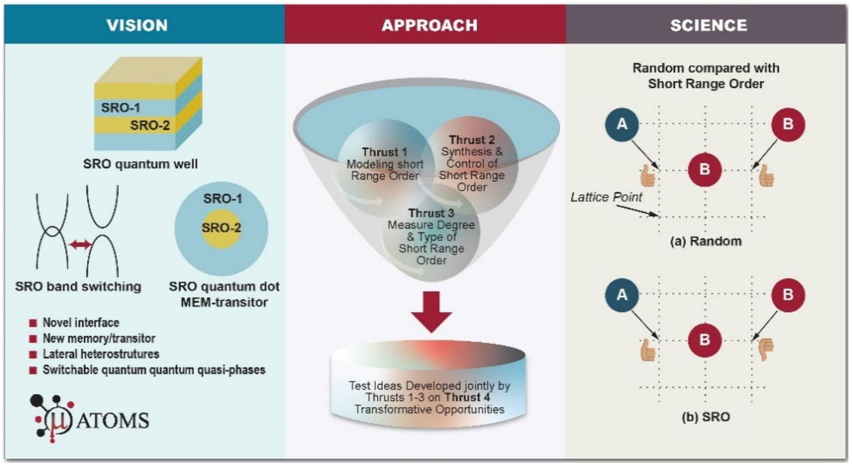

Figure 1 Vision: Precision manipulation of the spatial correlation among atoms as opposed to composition or dopants, to fabricate semiconductor properties using only one composition. **Approach:** Thrust 1-3 integrated into crosscutting Thrust 4 to demonstrate ability to design semiconductor properties and novel structures creating transformative opportunities. **Science:** SRO (a) Random – no restriction on nearest neighbors; (b) SRO - restriction on nearest neighbors.

The mission of the μ-ATOMS Energy Frontier Research Center is inspired by much of the work the research team recently uncovered on novel Group IV alloy semiconductor materials, demonstrating that short-range order (SRO) of Ge and Sn atoms on the periodic lattice of GeSn has a very large effect on the electronic energy band gap. SRO describes the probability that the nearest neighboring atoms to any B atom *(Figure 1)*, are restricted on average to NOT be the expected ratio of atoms A to atoms B, in the alloy A_xB_{1-x}. μ-ATOMS is aimed at uncovering the physics determining SRO, knowledge of the existing role of SRO on semiconductor properties, and new techniques to manipulate and control SRO. This is a knowledge base that can enable a scaled-up, reliable, and cost-effective sequence of operations for manufacturing semiconductor structures and devices from the bottom-up.

The impact is a new science of deterministic positioning, with a codesign purpose to utilize the spatial arrangements of atoms utilize spatial arrangements of atoms to synthesize novel heterostructures and graded morphologies using a single material, purposely ordered to form *(Figure 1)*: ***(i)*** SRO of atoms that defines low loss quantum wells, wires, dots, and corresponding photonic functions; ***(ii)*** different configurations of SRO that can be switched from one to the other as memory elements that truly converges transistors and memory, enabling facile in-memory computing and drastically increased energy efficiency; ***(iii)*** GeSn as a topological quantum material with SRO that harnesses and controls topological states for low-loss electrical transport; ***(iv)*** SRO of defects, such as vacancies, to lower electron-phonon scattering; and ***(v)*** domains of SRO that impact thermal transport much more than electrical transport, creating new opportunities for heat to electricity conversion. It presents opportunity for new microelectronic technology, leveraging a fundamental understanding of the underlying physics of the ordering of atoms in crystals, for new science on a silicon platform.

This mission will be realized by a team of researchers who are currently leading the experimental and modeling research breakthroughs on Group IV GeSn and SiGeSn semiconductor materials and who seek the underlying physics and chemistry principles for the ordering of atoms in semiconductor alloys as the knowledge base for a new technique for the synthesis for semiconductor properties and structures. The approach is rooted in a "Center Structure of Four Integrated Thrusts" *(Figure 1)* which parallel our four EFRC goals. The integrated thrusts and research goals of the Center are:

1. Model both material and structure to guide fabrication, and measurement of SRO in Group IV semiconductors. We are building on our early modeling results to precisely arrange Group IV (Si, Ge, Sn, Pb) crystal structures to discover and predict ***(i)*** the type and degree of SRO, ***(ii)*** the dependence on growth parameters, and ***(v)*** the expected performance of functional structures, such as QWs, using SRO.

2. Develop new synthesis tools and techniques to control and prepare Group IV semiconductors with different types and degrees of SRO. We are utilizing two different fabrication methods, spontaneous and stimulated, to achieve growth of different degrees of SRO and control over the spatial arrangement of SRO domains. Both methods rely on our pioneering new spontaneous and stimulated growth techniques, such as, amorphous crystallization, and Atomically Precise Advanced Manufacturing.

3. Explore new characterization tools to determine the ability to measure the type and degree of SRO. For example, we are leveraging our results, such as, using atomic probe topography and new simultaneous scanning transmission electron microscopy and Raman capabilities, to reveal the degree and type of SRO. We are using these tools to determine the correlation between SRO and material electrical and optical properties which are compared with our modeling predictions.

4.Control SRO in Group IV semiconductor alloys for transformative opportunities. We are determining the ability to control the type and degree of SRO in Group IV SiGeSnPb crystals by applying what we learn in thrust 1-3 to co-design and synthesize novel functional structures *(Figure 1)* This includes synthesizing controlled spatial arrangements of SRO domains to co-design quantum wells, wires, dots, waveguides, lasers, and transistors with memory.

The outcome from the μ-ATOMS EFRC is to demonstrate fundamental understanding and precision manipulation of SRO in Group IV semiconductor alloys as a new tool and to deliver new science and novel device concepts on a silicon chip.

μ-ATOMS	
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Sandia National Laboratory	Ezra Bussmann, Shashank Misra, Tzu-Ming Lu, Michael Lilly
George Washington University	Tianshu Li
Rensselaer Polytechnic Institute	Shengbai Zhang
Dartmouth College	Jifeng Liu
Stanford University	Paul McIntyre
University of California Berkeley	Andrew Minor
Arizona State University	Yong-Hang Zhang, Dragica Vasileska
University of Delaware	Yuping Zeng
University of Arkansas at Pine Bluff	Mansour Mortazavi

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Ensembles of Photosynthetic Nanoreactors (EPN)
EFRC Director: Shane Ardo
Lead Institution: University of California Irvine
Class: 2022 – 2026

Mission Statement*: To understand, predict, and control the activity, selectivity, and stability of solar water splitting nanoreactors in isolation and as ensembles.*

The overarching question that guides the scientific mission of **EPN** is: *How can the solar-to-hydrogen energy conversion efficiency of ensembles of photosynthetic nanoreactors be increased by more than one order-of-magnitude*? To answer this question, **EPN** is taking a bottom-up approach to synthesis, characterization, and modeling of solar water splitting nanoreactors. Fundamental knowledge gained will be used to identify the physicochemical principles that in aggregate are responsible for ensemble behaviors. This information will guide **EPN** toward achieving its scientific mission, and may lead to transformative pathways to meet the DOE H_2 Earthshot cost target of $1 per kg-$H_2$.

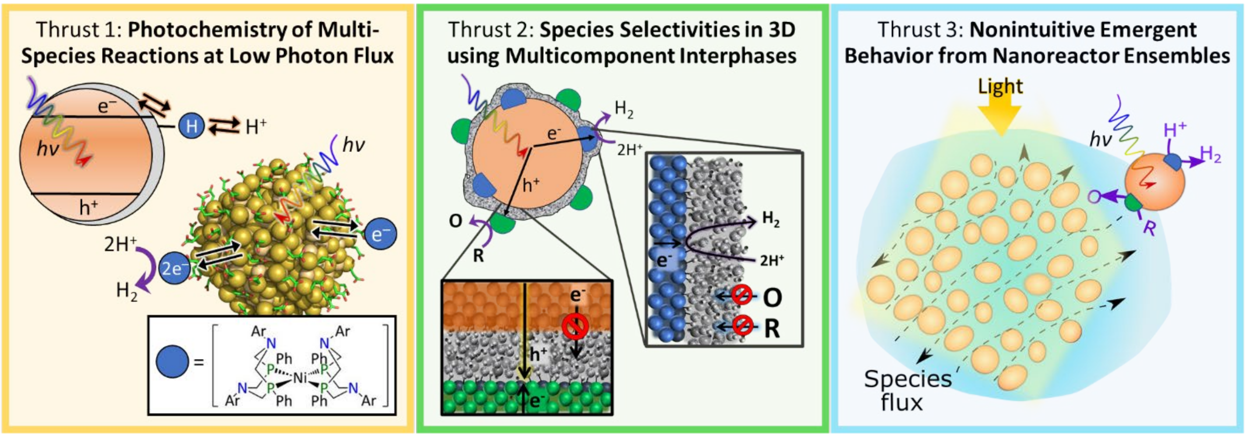

Fig. 1 | **EPN**'s multifaceted research thrusts aimed at unveiling design rules for solar water splitting nanoreactors.

EPN's three thrusts shown in *Fig. 1* benefit from integrated efforts and close collaborations in nanomaterials synthesis, advanced microscopies and spectroscopies, and multiscale physics-based and data-driven computational modeling. Building on knowledge gained from planar model systems, synthesized nanoreactors will be studied in isolation and as ensembles containing several-to-millions of nanoreactors. Outcomes will inform codesign strategies for the four interacting microenvironments critical to **EPN**: *(i)* the semiconducting solid phase; *(ii)* multicomponent electrocatalytic interphases; *(iii)* the intervening aqueous liquid phase between adjacent nanoreactors; and *(iv)* the collective terrestrial and solar blackbody radiation fields. By controlling properties of these four microenvironments, **EPN** will reveal means to achieve high quantum yields and energy conversion efficiencies for each elementary step, from transport of incident solar photons to formation of chemical products. Because each nanoreactor inherently cogenerates both oxidation and reduction reaction products in close proximity, exquisite control over species selectivities is paramount. A core hypothesis within **EPN** is that reaction selectivities can be controlled independently for electrons, holes, reactants, and products, through precise molecular-level design and synthesis of multicomponent interphases. To better understand how ensemble behaviors arise from elementary steps, **EPN** will also evaluate reactivity from smaller, well-defined ensembles of interacting nanoreactors. This is important, because simulations indicate that solar-to-hydrogen energy conversion efficiencies for ensembles of photosynthetic nanoreactors can exceed those of standard photoelectrochemical devices. This outcome arises from the low absorbed photon flux per nanoreactor, resulting in small photocurrents and small overpotentials, combined with the multiplicative output of having many nanoreactors in an ensemble.

Inherent to the naturally interwoven aspects of **EPN** is the need for a convergent multidisciplinary center-scale effort with diverse and synergistic theoretical and experimental capabilities. Nanoreactors containing multicomponent functional interphases will be characterized using a correlative microscopy platform that aligns experimental microscopic and spectroscopic capabilities across multiple complementary techniques to quantify underlying properties of single nanoreactors. Generally, it remains unknown whether experimentally measured ensemble activity is dominated by several high-efficiency nanoreactors or a few nanoreactors that serve as catastrophic shunts. This will be revealed through development and use of cross-platform-compatible liquid microscopy cells and light excitation sources that allow for identical-location *in situ* correlative microscopic characterization of individual nanoreactors exhibiting varying performance. Experimental observations will be interpreted using a multiscale simulation network, which connects modeling expertise across a multitude of length and time scales to simulate the interplay of optical, species, and thermal processes. Theoretical models of stochastic and ensemble processes will be refined based on outputs from atomistic/molecular-level simulations. With experimentally validated physics-based predictions for ensemble performance, data-driven machine-learning models will be used to solve the inverse problem of designing nanoreactors to achieve desired ensemble performance metrics. These links are crucial to bridging knowledge gaps for the influence of multicomponent functional interphases on ensemble reactivities, leading to desired redox selectivities. Furthermore, knowledge gained from **EPN** will provide guidance to research and development of batteries, fuel cells, membranes, and other photochemical devices, each that benefit from atomic-level control over functional interphases.

EPN includes diverse minority representation (AANAPISI, ANNH, HSI, NASNTI, PBI/HBCU) and primarily undergraduate institutions (PUIs). It is organized into three clusters by geographic location (California, Colorado, Northeast) to foster regional collaborations and a culture of camaraderie. To complement **EPN**'s world-class innovative research, **EPN** is developing a program aimed at training the next generation of scientists and engineers interested in microscopy, while strengthening the STEM pipeline between PUIs and R1 institutions through mutually beneficial research partnerships. Each PUI is intentionally situated near an R1 institution to help facilitate exchange of R1 graduate students and PUI undergraduate students between institutions.

Ensembles of Photosynthetic Nanoreactors (EPN)	
University of California Irvine	Shane Ardo (Director), Nien-Hui Ge, Xiaoqing Pan, Joseph Patterson, Jenny Yang
California Institute of Technology	Scott Cushing
California State University, Long Beach	Young-Seok Shon
City University of New York, Medgar Evers College	Oluwaseun Salako
Colorado State University	Justin Sambur
Columbia University	Daniel Esposito
Fort Lewis College	Kenneth Miller
Lawrence Livermore National Laboratory	Tadashi Ogitsu
National Renewable Energy Laboratory	Katherine Hurst
Sandia National Laboratories	Albert Talin
University of Colorado Boulder	Gordana Dukovic
University of Michigan	Rohini Bala Chandran
Yale University	Shu Hu

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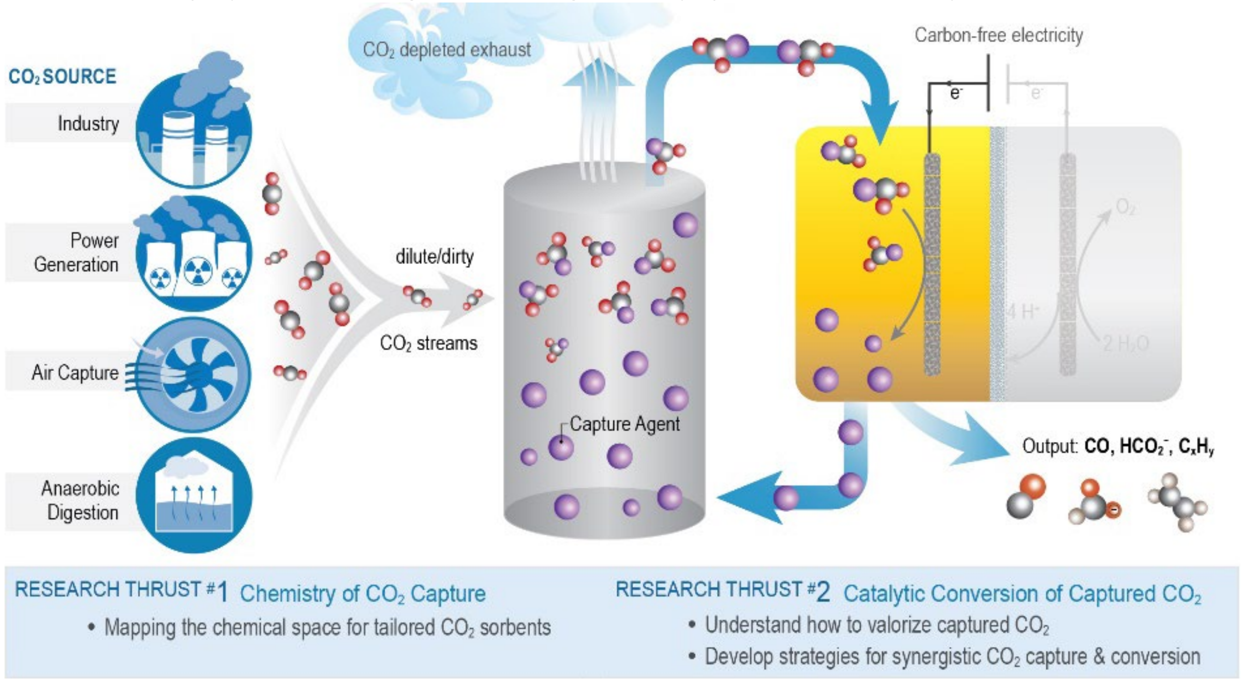
Center for Closing the Carbon Cycle (4C)
EFRC Director: Jenny Y Yang
Lead Institution: University of California, Irvine
Class: 2022 – 2026

Mission Statement*: To advance synergistic capture and conversion of carbon dioxide (CO_2) from dilute streams into useful products through the convergent study of sorbents and catalysts.*



The **Center for Closing the Carbon Cycle (4C)** will advance the foundational science and define key integration parameters for synergistic CO_2 capture and conversion, or reactive capture of CO_2 (RCC). While there has been significant independent research in either CO_2 capture or pure CO_2 conversion, it is not well understood what knowledge is translatable to RCC. 4C combines expertise on capture and valorization in center-wide collaborations what will enable co-design of CO_2 sorbents with catalysts for conversion. 4C will establish guidelines for CO_2 capture from various dilute and dirty streams and define how captured CO_2 can most effectively be utilized, leading to selective, durable, and efficient pathways from CO_2 source-to-product.

Research Thrust #1 (RT #1) is focused on the chemistry of CO_2 capture. Libraries of CO_2 sorbents will be established using computational screening in parallel with high-throughput experimentation. These studies will establish structure-electronic relationships for different classes of soluble sorbents and functionalized solvents and their stability towards oxygen, water, and other common contaminants. The effect of electrolyte and other additives on sorbent properties will be quantified, and their microstructures will be spectroscopically and computationally interrogated. Neutron scattering will provide a link between macroscopic CO_2 binding and atomistic structure, and picosecond dynamic simulations will provide crucial insights to speciation and structural reorganization at atomic length scales. This information be used to develop improved models for CO_2 binding, solvation energies, and microenvironment effects on sorbent molecules and materials.

The research will advance our understanding of CO_2 sorption chemistry and expand the library of CO_2 sorbents. The knowledge base will valuable for understanding how to capture CO_2 from air or other point sources, which is increasingly important for carbon neutral and net negative technology outside of RCC.

Research Thrust #2 (RT #2) will focus on electrocatalytic valorization of captured CO_2. Our understanding of mechanisms and guiding concepts in electrochemical CO_2 Reduction (CO2R) has grown to include canonical reaction paths and thermodynamic descriptors for reactivity and selectivity. The discovery of new catalysts and electrolyte interactions for CO2R have illuminated effects of specific ions, dielectric constant, and pH on activation barriers, reaction selectivity, and product selectivity. 4C aims to understand how these principles translate to RCC, as captured CO_2 adducts are an intrinsically different substrate for catalysis with a broad range of molecular diversity, as described in RT #1. RCC allows tuning the physical and reactivity properties of the substrate, captured CO_2, to access a broader scope of chemical transformations compared to traditional CO_2 chemistry. Moreover, the dynamics between CO_2, the capture agent, supporting electrolyte, and solvent create new catalyst considerations.

4C will build on our understanding of electrochemical CO2R by establishing the concepts that underpin the reactivity of captured CO_2. By bridging knowledge between capture and conversion, 4C will enable the co-design efficient and selective homogeneous, heterogeneous, and hybrid catalysts for RCC to C-based commodities and fuels. A key theme is understanding the interface between substrate, solvent, and electrolyte with catalyst active sites, which is necessary to improve models and develop new theories regarding the design of catalyst microenvironments. These principles are broadly applicable, and will be transformative in how the community considers the role of interfaces and chemical microenvironments.

Summary. By studying CO_2 capture (RT #1) and conversion (RT #2) together, 4C will address the fundamental challenges associated with integration, including identifying and matching the relevant kinetics at multiple time scales to enable continuous operation. In addition to improving the overall efficiency and thus lowering the cost of CO_2-derived products, cooperative research between capture and conversion will lead to systems that more economically valorize CO_2 from dilute and dirty streams.

Center for Closing the Carbon Cycle (4C)	
University of California, Irvine	Jenny Yang (Director), Robert Nielsen, Vy Dong
California State Polytechnic University, Pomona	Chantal Stieber
California Institute of Technology	John Gregoire
Case Western Reserve University	Burcu Gurkan
Elizabeth City State University	Bijandra Kumar
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University of Colorado, Boulder	Wilson Smith
University of Louisville	Joshua Spurgeon
University of Michigan	Charles McCrory

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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 – 2024

***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 this mission, the SCALAR-EFRC has defined five scientific objectives that we feel represent particularly promising routes to these goals. As a center, we aim to:

1) The overarching center goal is to take a multiscale view of materials design and fabrication, using diverse synthetic methods to optimize battery materials from the micro- to the macro-scale.

2) Thrust I specifically aims to improve energy density through the development of insertion hosts that show reversible multi-electron redox.

3) Thrust II focuses on improving power density by facilitating ion and electron transport using both atomistic and architectural optimization.

4) Thrust III takes on the challenge of improving reversibility through the optimization of bulk and interfacial structures, focusing on the intertwined goal of improving both chemical and mechanical stability.

5) A holistic view of all these efforts can be found in the overarching goal of reducing hysteresis in all aspects of electrochemical energy storage.

Within these goals, Thrust 1 aims to increase electrode capacity using a combination of anion intercalation chemistry, mixed anion and cation redox in extended inorganic solids, and two electron cation redox. Thrust 2 attacks the problem of improving power density by facilitating both electron and ion mobility in battery materials. This thus effort combines conjugated polymers as binders with increased electrical and ionic conductivity, high ionic mobility phases, intrinsically conductive electrode materials, and nanoscale architectures. These efforts are coupled with electrochemical calorimetry to directly measure thermal loss within our systems. Finally, Thrust 3 aims to solve the challenge materials stability using inorganic surface coatings for improved stability and reversibility, flexible nanoporous architectures that can mitigate and even compensate for large volume changes during cycling, and catalysts that can favor desired electrochemical reactions and mitigate unfavorable ones.

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 our understanding of battery materials and have long term impact on how we think about the kinds of new materials and reactions that can be utilized for rechargeable electrical energy storage.

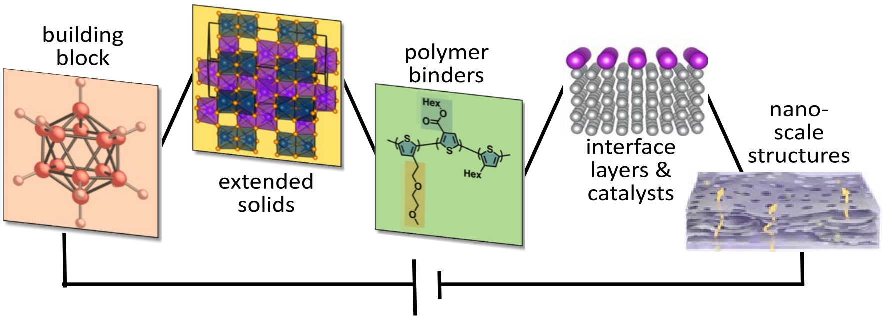

Figure 1: The SCALAR EFRC aims to rethink battery materials, starting from the level of building blocks and then moving to new motifs for extended solids. New materials are integrated with multifunctional polymer binders, designer interfaces, and controlled nanoscale architecture.

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 (Assistant Director), Xuan Duan (Thrust 3 Lead), Laurent Pilon, Philip Sautet
University of California, Santa Barbara	Ram Seshadri (Thrust 2 Lead), Bradley Chmelka, Anton Van der Ven
University of Southern California	Brent Melot (Thrust 1 Lead), Sri Narayan, Barry Thompson
Caltech	Kimberly See
Stanford Synchrotron Radiation Lightsource	Joanna Nelson Weker
University of California, San Diego	Jian Luo

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Quantum Materials for Energy Efficient Neuromorphic Computing (Q-MEEN-C)
EFRC Director: Ivan K. Schuller
Lead Institution: UCSD
Class: 2018 – 2026

***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 a brain ("neuromorphic").*

The digital and computational revolution of the last seven decades has propelled arguably one of the most important technological advances in history. The key breakthrough was the realization that a material such as Silicon features electronic properties that can be incorporated into a transistor, the basic building block of a von-Neumann computer. The current vertiginous development of storage density, computational power and cost is approaching the end due to unsustainable local high energy demand. The key future requirement of a reliable computation architecture, aside from being fault-tolerant, is to be energy efficient. The brain performs complicated computational tasks with high energy efficiency. The human brain serves as an inspiration to emulate this behavior using materials that can mimic dendrites, synapses, neurons, axons, and the network they comprise.

The original Q-MEEN-C approach included two materials-focused Thrusts: Charge and Spin. Since, in quantum materials, the two phenomena are strongly linked and can influence each other, we have reconfigured the project into 3 closely integrated materials-focused Thrusts **(Fig. 1)**. The principal nanoscale Quantum Materials (Thrust 1) will test and feedback the Neuromorphic Functionalities (Thrust 2) occurring at the mesoscale and use and modify the Emergent Phenomena (Thrust 3) which arise in macroscopic systems because of their coupling.

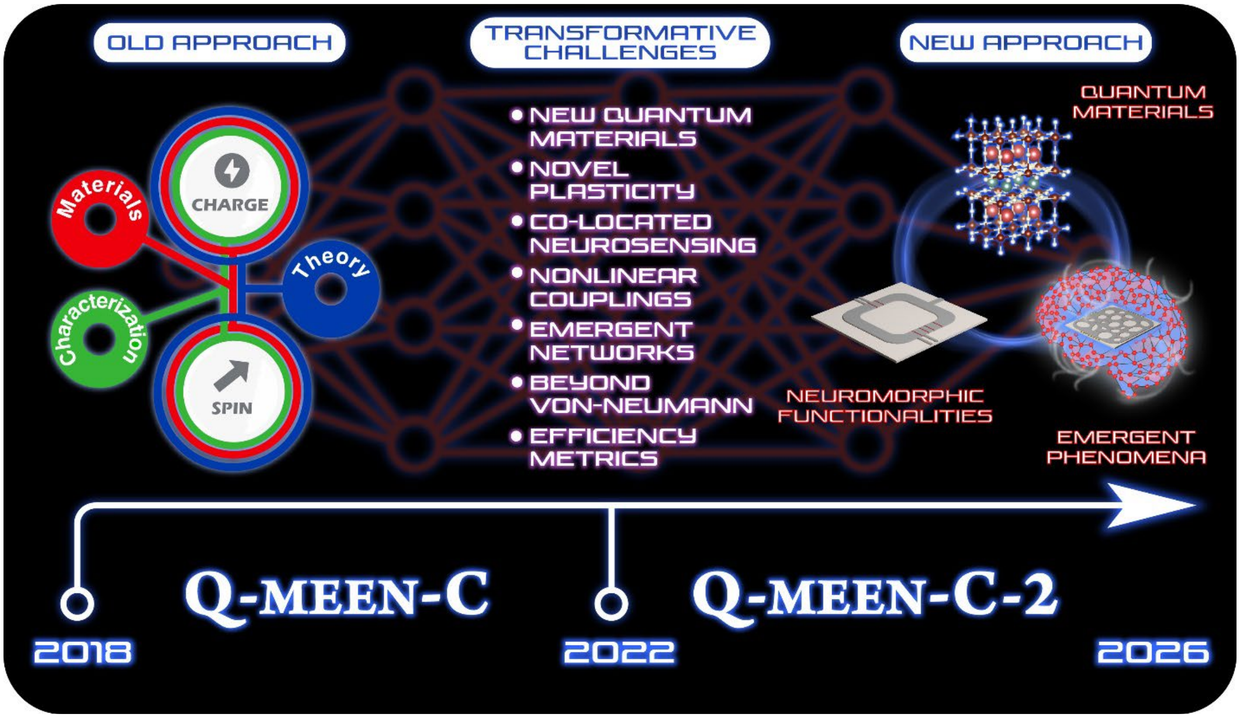

***Figure 1:** Based on the accomplishments in the first 4 years with Spin and Charge thrusts, we identify 7 transformative challenges which lead to our newly structured, three-pronged approach.*

During the next 4 years Q-MEEN-C will synthesize promising new materials candidates, understand their microscopic and mesoscopic behavior due to naturally occurring and/or artificially imposed inhomogeneities, develop novel contactless connectivity using collective or frequency selective mesoscopic coupling, and develop new performance benchmarks for important materials properties. While the project is "materials-centric", its influence goes beyond the development and understanding of materials at the nanoscale to the meso- and macroscale properties.

This project is dedicated to fundamental research on materials that will mimic key properties of the brain, to develop a brain-inspired, "neuromorphic" computational paradigm. We will study aspects of materials at hierarchical length scales, from the microscopic to the macroscopic, in the same way that the brain itself is understood as an emergent phenomenon **(Fig. 2).** Central to our research is the realization that "quantum materials", which feature richer and more complex properties than conventional semiconductors, can offer emergent properties parallel to those of the brain.

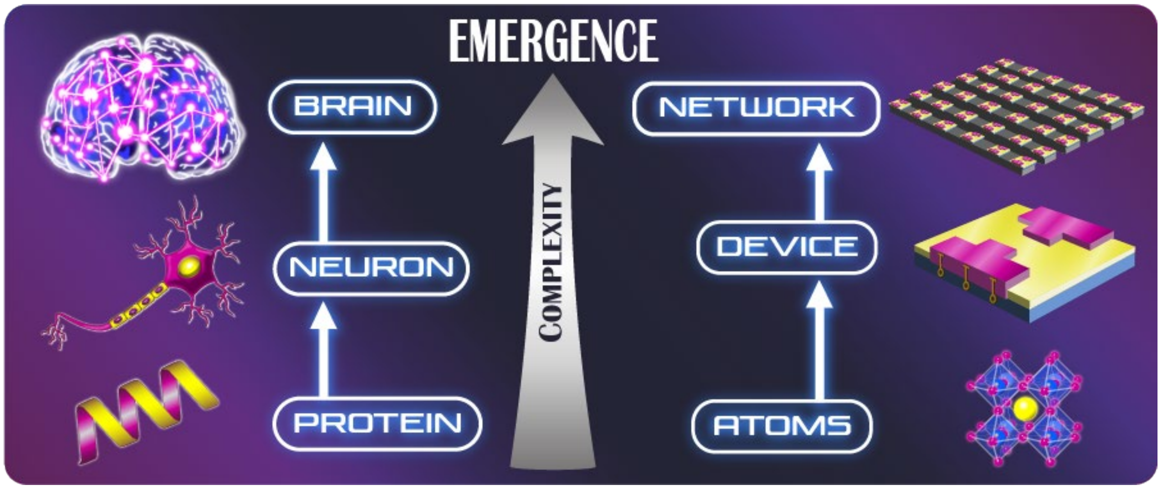

Figure 2: *We mimic the emergent complexity that makes the brain an efficient computer by harnessing the properties of quantum materials, approaching the problem at various length scales.*

Quantum Materials for Energy-Efficient Neuromorphic Computing (Q-MEEN-C)	
UC San Diego	Ivan K. Schuller (Director), Oleg Shpyrko, Alex Frañó, Robert Dynes, Eric Fullerton, Marcelo Rozenberg, Duygu Kuzum
UC Davis	Yayoi Takamura
UC Santa Barbara	Jonathan Schuller
NY University	Andrew Kent
Rutgers University	Shriram Ramanathan
University of Chicago	Giulia Galli
University of Illinois, Urbana-Champaign	Axel Hoffmann
CNRS, France	Julie Grollier
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Brookhaven National Lab	Yimei Zhu

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EFRC Catalyst Design for Decarbonization Center (CD4DC)
EFRC Director: Laura Gagliardi
Lead Institution: University of Chicago
Class: 2022 – 2026

***Mission Statement**: To discover and develop reticular metal-organic framework materials as catalysts for the decarbonization energy transition and to optimize the key catalytic reactions involved.*

The call to decarbonize the chemical and energy industries requires the reduction and eventual elimination of fossil carbon resources and the adoption of radically new approaches for producing chemicals and storing electric power harvested from the wind and sun. To respond to this call, the central research mission of CD4DC is to discover and develop reticular metal-organic framework materials (MOFs) as catalysts for the decarbonization energy transition and to optimize the key catalytic reactions involved. This will be accomplished by a symbiotic combination of synthesis, catalysis and kinetics, computational modeling and active learning, and characterization.

Using widely abundant hydrogen as a primary energy vector would reduce the need for carbon rejection and replace it with hydrogen addition, which requires comparatively lower operating temperatures that will reduce the overall carbon footprint. Future approaches must include processes that convert electric power to chemical energy, and the initial transformation of electric energy to chemical energy will involve electrolysis and generation of H_2. A large percentage of hydrogen will be intermittently stored in liquid organic hydrogen carriers (LOHCs) that can release H_2 at the time and place of the target application. The extraction reaction is endothermic, and its utility will depend critically on a combination of the efficiency of dehydrogenation and separation of the dehydrogenated carrier and hydrogen. These processes require highly precise, robust, and stable catalysts for the key generic transformations, namely the addition and release of hydrogen to and from organic molecules and the manipulation of C-C bonds. Catalysts with these desired properties that operate with high efficiency and at low temperatures are not currently available.

CD4DC will target the development of three types of catalysts: (1) MOFs with components that provide a high degree of polarizability and softness for superior hydrogen transfer catalysis, (2) MOFs that enable the use of an external electric potential to facilitate catalytic transformations, and (3) MOFs that provide bio-inspired environments for highly selective chemical transformations. Project objectives will be achieved through interactive and iterative efforts of synergistic computational and experimental techniques that leverage our unique, multi-disciplinary team. These themes will systematize the design of catalysts that drive the two proposed reaction classes: H_2 addition and release from LOHCs, and the efficient combination of C–C bond formation with H_2 management.

MOFs are the central component of the design strategy because of their unmatched diversity within the boundaries of a well-defined class of materials, the uniformity of their characterizable active centers, the tailorable sterically and chemically well-defined environments near the active site, and the ancillary components that modulate transport and selectivity. These properties facilitate explanatory and exploratory molecular-level computational modeling of catalyst properties for molecular-level control of catalytic transformations. The theoretical and computational component of our center, aided by active learning, will identify key structure–function relationships for catalytic activity and aid hypothesis-driven understanding and design of catalysts and catalytic pathways. Such design—particularly of the catalytic active center, environment, and reaction mechanism—creates a synergistic cycle between theory, experiment, and active learning that will drive success.

Our choice of target reactions addresses the emerging challenges in catalysis science for decarbonization. As society seeks to transition from C-centered energy and industrial schemes, processes that enable efficient addition and removal of H_2 will become increasingly important. Therefore, we will investigate the fundamental mechanisms of (bio)alcohol dehydrogenation to aldehydes and aldehyde oligomers and hydrogen addition and removal from aromatic LOHCs. The move away from a petroleum-based economy will require selective, low-energy processes for generating higher-order chemical feedstocks from biogenic sources or recycled waste. We will also examine reaction mechanisms for C–C bond formation, such as the coupling of CO, carboxylates, or alcohols, which also will require close management of H_2. A particular challenge is to realize C–C coupling reactions (i.e., ones involving H_2 addition) with high selectivity. Highly active and selective versions of these reactions could be truly transformative for decarbonization of the chemical industry.

At the end of the four-year program, we will have laid the foundation for the decarbonization transition by understanding and validating the requirements for targeted chemical transformations using MOFs and will have advanced the tools for de novo design of catalysts via modular synthesis. Moreover, the active learning-driven integration of high-throughput experimentation and computation will generate significant new knowledge on MOF-based catalysis that will be available to the broader energy research community.

Catalyst Design for Decarbonization Center (CD4DC)	
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Argonne National Laboratory	Massimiliano Delferro
Clemson University	Rachel Getman
Northwestern University	Omar Farha Joseph Hupp (Deputy Director) Justin Notestein
Pacific Northwest National Laboratory	Johannes Lercher Nancy Washton (Deputy Director)
Stony Brook University	Karena Chapman
University of Illinois at Chicago	Ksenija Glusac
University of Minnesota	Matthew Neurock Donald Truhlar

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

Mission Statement*: To: (1) Develop catalytic and functionalization approaches and fundamental tools applicable to the upcycling, upgrading, and recycling of real polymer plastics waste (PPW) with a strategic focus on enabling mixed-stream transformations in varied material forms (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.*

The ***Center for Plastics Innovation (CPI)*** was launched in August 2020. Led by the University of Delaware (UD), *CPI* brings together researchers from UD; the University of Chicago; the University of Massachusetts, Amherst; the University of Pennsylvania; and Oak Ridge National Laboratory (ORNL) to address the global challenge of transforming polymer plastics waste (PPW) into valuable products. 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. *CPI* is organized into three synergistic thrust areas (**Figure 1**). *Thrust 1: PPW Deconstruction* focuses on converting PPW into fuels, lubricants, precision macromolecules, and monomers *via* tunable heterogeneous catalysis with a focus on selectivity, scalable, and lower-energy approaches. *Thrust 2: PPW Upcycling* develops functionalization and upcycling strategies using plasma-assisted catalysis and surface treatment, photo-redox catalysis, enzymatic and microbial routes to generate high-value monomers and polymers. *Thrust 3: Cross-cutting Tools* enables *CPI's* recycling/upcycling/upgrading advances *via* artificial intelligence (AI) strategies, computational modeling activities, additive manufacturing approaches, and macromolecular characterization.

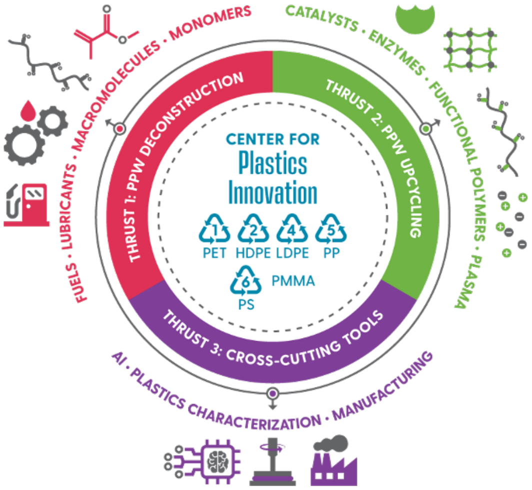

Figure 1. CPI research thrusts.

Background and Knowledge Gaps: Plastics play an indispensable role in every aspect of modern life. Globally, the manufacture of plastic products was ~311 million tons in 2014, and that number is estimated to double by 2034. Increased plastics production also exacerbates the projected environmental impacts (*e.g.,* landfill usage, aquatic pollution, degradation byproducts), as in the U.S. less than 10% of polymer plastics waste (PPW) is actually collected for recycling. The increasing demands for lightweight and resilient materials, along with the significant environmental threats from plastics waste and pollution, particularly from single-use plastics, require a new paradigm in end-of-life plastics management. We will leverage our expertise in catalysis (synthetic, biological), macromolecular science and engineering, additive manufacturing, data science and 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.*

Impact: We will transform the current high-energy/low-value landscape of polymer recycling towards highly efficient polymer upcycling strategies. Mechanistic insights in low-temperature catalysis for polymer deconstruction in the melt, plasma-assisted functionalization approaches, photoredox strategies for chemical transformation, and enzyme engineering to valorize PPW will provide immense fundamental knowledge. Cross-cutting experimental, computational modeling, and data science 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. Our comprehensive program has the **overarching goal** to overcome the fundamental knowledge barriers described above toward advancing PPW chemical recycling and upcycling strategies. The **goals** are to: (1) Develop design principles for catalysts that deconstruct mixed PPW at low temperatures, avoid coking, are resilient to impurities, and produce tunable distributions of products; (2) Design hierarchical, multiscale catalysts to enable fast heat and mass transfer and methods to disperse catalysts in plastics to increase energy efficiency during deconstruction processes; (3) Develop multiscale methods and experimental methods to predict and measure physical property data for polymer processing; (4) Reveal and predict interactions and chemical transformations of macromolecules in the melt, on catalyst surfaces, and in confined spaces; (5) Develop functionalization methods of PPW-deconstruction products through plasma-catalyst interactions, photoredox-catalyzed reactions, and biocatalysis to enable upcycling and recycling; (6) Synthesize and characterize reprocessable, high-performance network polymers; (7) Develop enabling tools, such as AI, multiscale models, 3D-printing and additive manufacturing capability, and an integrated molecular-systems analysis framework to enable catalyst discovery, efficient chemical recycling, and plastics upcycling; (8) Educate the U.S. workforce; and (9) Enable technology transfer.

Center for Plastics Innovation (CPI)	
University of Delaware	LaShanda Korley (Director), Thomas Epps, III (Deputy Director), Michael Berg (Science & Technology Director), Mark Blenner, Stavros Caratzoulas, Wilfred Chen, Hui Fang, Emil Hernandez-Pagan, Laure Kayser, Aditya Kunjapur, Raul Lobo, Michael Mackay, Eleftherios Papoutsakis, Joel Rosenthal, Kevin Solomon, Dion Vlachos, Mary Watson
Oak Ridge National Laboratory	Josh Michener
University of Chicago	Juan de Pablo, Stuart Rowan
University of Massachusetts, Amherst	Omar Abdelrahman, Peng Bai
University of Pennsylvania	Raymond Gorte, Daeyeon Lee

Contact: LaShanda Korley, Director, lkorley@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 – 2026

***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 their molecular spin states and their coupling to external fields and the environment. The objective of the Center for *Molecular Magnetic Quantum Materials* **(M^{2}QM)** is to discover, develop, and deliver the pivotal materials physics and chemistry knowledge of molecular magnetic quantum materials essential for quantum information technologies. M^{2}QM aims to transform molecular magnets (MMs) from promising building blocks into viable *quantum materials* that are useful both for coherent quantum information systems (QIS) and for quantum electron-spin devices.

M^{2}QM focuses on providing the crucial synthesis, characterization, aggregation and assembly, adaptation, and control needed for utilization of MMs as quantum materials. The central technological contexts are to develop technologies that enable quantum information processing for quantum computation, to impact problems critical to humanity, and to fulfil the potential for exquisite sensitivity and specificity of quantum sensing that utilizes quantum effects.

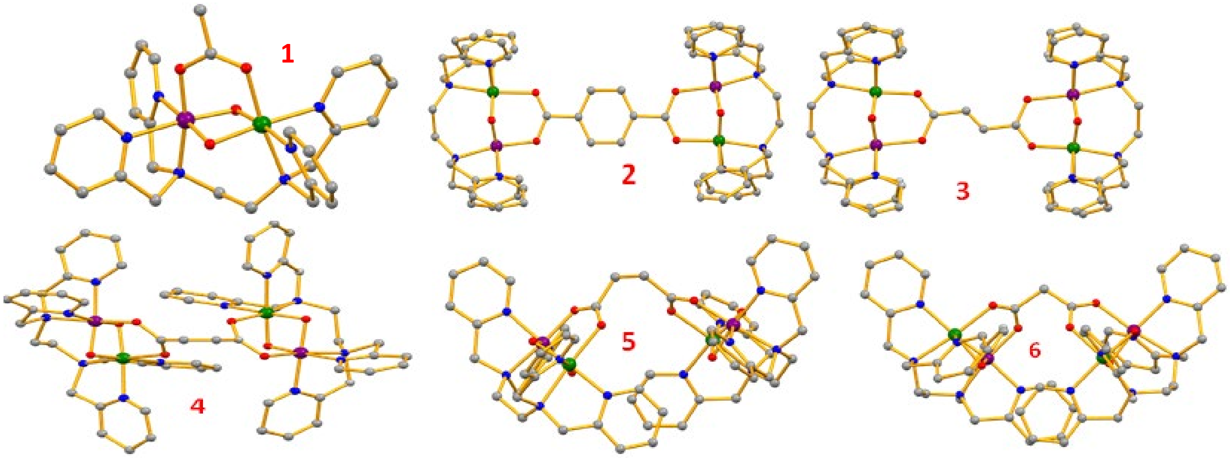

Fig.1 The [Mn$_2$] magnetic molecule with $S=1/2$ {1} and its conversion into dimers 2-6 by covalent linkage with a variety of dicarboxylates.

The Center provides a unique integration of advanced theory, innovative experiment, and expert chemical synthesis to design, probe, and control the quantum degrees of freedom of electron spins at the scale of molecules. Molecular spins offer the potential of almost limitless tunability via the techniques of coordination chemistry, with respect to coherence, energy gaps, and couplings. Furthermore, molecular qubits are completely identical and easy to create in large numbers. Figure 1 shows how a single spin center molecule can be linked chemically to create five different dimers. However, there remain many challenges for realizing quantum information (and other quantum) applications with molecules in practice. Those challenges primarily are related to control, addressing, and integration into technologies at larger scales while controlling decoherence.

M^{2}QM brings together experts in the synthesis of molecular magnets, characterization with unique spectroscopies, technological integration via surface and macromolecular chemistries, and computational and theoretical modeling of quantum properties, to carry out fundamental research to identify new and promising molecular platforms on which to make advances upon the specific challenges noted above.

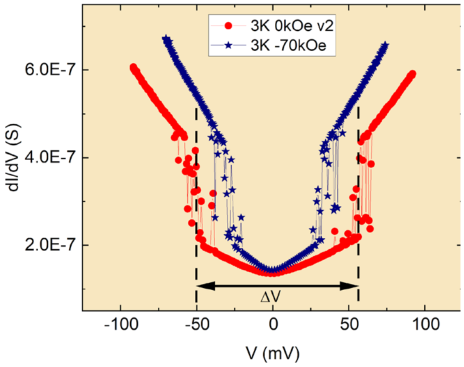

Fig. 2. Differential conductance plotted as a function of voltage bias shows field-dependent energy transitions between quantum states in an MnPc film.

We envisage three fundamental directions for the second M^{2}QM research term. First is to utilize the understanding of decoherence pathways gained in the first phase to design new MM systems with long coherence times. On those we will demonstrate two qubit gate operations at speeds that are fast compared to the coherence time. Second is to move from promising demonstrations of magnetoelectric couplings in bulk structures to a more focused design that exploits the relatively soft MM lattices and the tunability of individual MM spin configurations to achieve very strong magneto-electric coupling. Third is to demonstrate functional control of MM films and substrate interactions, for example, to achieve tunneling based control. All these directions will be supported by an intense theoretical and computational effort to simulate the quantum properties of individual molecules and aggregates, as well as machine-learning based discovery of new candidate molecules and aggregates. Figure 2 shows, for example, transport measurements of a MnPc thin film demonstrating transitions between quantum magnetic states are acquired using a 1 mV rms voltage modulation. A smaller voltage modulation should resolve additional energy levels in the transition regions.

M^{2}QM's quest to deepen and broaden the understanding of critical materials physics and chemistry of MMs for QIS is built upon accomplishments from the first funding period (EFRC class of 2018) and targeted at the fundamental science needed for eventual applications. That includes both quantum devices (qubits) and quantum-current (e.g., spin) systems. The strategy is pursued via three Thrusts, each an experimental-theoretical-computational team. Theory-computation efforts across the entire Center are enhanced via the Cross-cutting Theory-Computation team. M^{2}QM's bottom-up approach embodied in this organization is to synthesize MMs and linker molecules with high promise of properties useful for QIS, to achieve stable condensed assemblies of those molecules that preserve those desirable properties (chiefly spin-state control and coherence), and to demonstrate coupling, modulation, control, and coherence preservation required for QIS applications.

Center for Molecular Magnetic Quantum Materials (M^{2}QM)	
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Florida State University	Stephen Hill (associate director), Mike Shatruk
University of Central Florida	Talat Rahman
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Quantum Sensing and Quantum Materials (QSQM)
EFRC Director: Peter Abbamonte
Lead Institution: University of Illinois, Urbana-Champaign
Class: 2020 – 2024

Mission Statement: *To develop new quantum sensing techniques, scanning qubit microscopy and two-electron EPR spectroscopy, and apply them to investigate relationships between the local and global properties of quantum materials that exhibit strong correlations and/or topological order: exotic superconductors, topological crystalline insulators, and strange metals.*

The EFRC on Quantum Sensing and Quantum Materials (QSQM) will apply new types of quantum sensing to investigate the local and nonlocal quantum observables in three families of quantum materials central to DOE's energy mission: exotic superconductors, topological crystalline insulators (TCIs), and strange metals. Each family presents a compelling mystery: the *origin of chiral or topological behavior* in exotic superconductors, whether *topologically protected corner and hinge states exist in new generations of TCIs*, and the *origin of Planckian dissipation* in strange metals. The two quantum sensing techniques we develop will enable us to unravel the mysteries of these phases in quantum materials:

1. *Scanning qubit microscopy* to investigate localized electronic states and fluctuations.
2. *Two-electron EPR spectroscopy* to investigate electron correlations and interaction mechanisms.

For the first, we are developing a **scanning qubit microscope (SQM)** (Fig. 1), which consists of a single flux qubit integrated into a scanning resonator tip that can be translated over the surface of a material. The state of a qubit is highly sensitive to charge and flux noise, making an SQM a sensitive probe of local charge and spin fluctuations near surfaces or interfaces. A qubit is therefore a bulk-sensitive, non-contact, non-destructive probe that can study low-energy excitations in materials an with unprecedented resolution of 10 kHz. A qubit can detect both electric and magnetic fields, and can be used for quantum manipulation, allowing us swap excitations between the qubit and the material. This SQM will elucidate localized defects in exotic superconductors, fluctuations in strange metals, and localized excitations and boundary modes in TCIs.

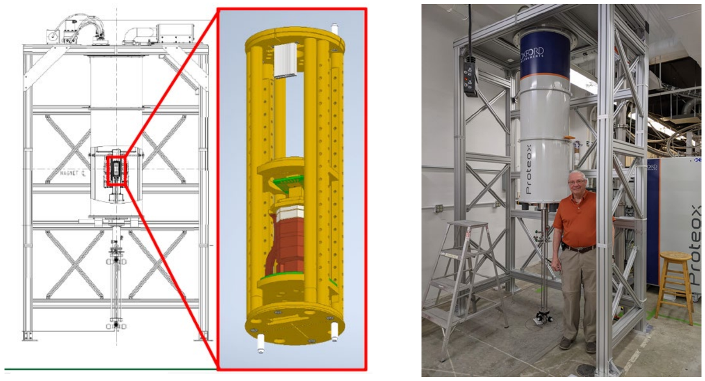

Fig. 1: Conceptual layout of a scanning qubit microscope. (left) Dry dilution refrigerator, sensor head, and scanner. (right) Dedicated SQM setup recently installed in the Materials Research Laboratory at the University of Illinois.

For the second technique, we are developing a **two-electron Einstein-Podolsky-Rosen (EPR) spectrometer** (Fig. 2), which uses correlated EPR pairs of electrons to reveal valence band interaction effects. This instrument can be operated either with an ultrafast XUV laser source or with a pulsed ultrafast electron source. The former allows one to coherent pairs of photoelectrons (a technique known as "double photoemission"), revealing the correlation between pairs of electrons in a material. The latter allows one to study scattering between pairs of electrons (at technique known as "Rutherford scattering"), revealing the renormalized interaction mechanism between the electrons. This instrument will therefore reveal a complete picture of the Cooper pairing mechanism in exotic superconductors, the microscopic origin of electron-electron interactions and the degree of correlations in strange metals and TCIs.

Using these techniques, as well as mature approaches in materials synthesis and characterization, the QSQM will be able to address the following profound materials problems:

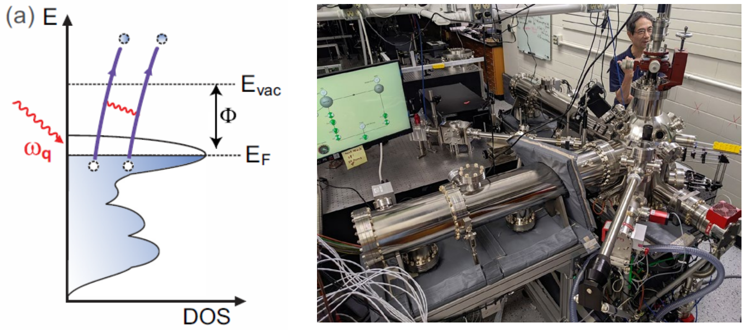

Fig. 2: EPR spectrometer under commissioning in the Materials Research Laboratory at the University of Illinois. (left) Illustration of double ARPES, which reveals how electrons are correlated in a material. (right) New setup showing two ARTOF-2 time-of-flight analyzers, which are run in coincidence to identify correlated EPR electron pairs.

Exotic superconductors: We aim to identify the pairing bosons and origin of broken symmetries and topological order in exotic superconductors, including chiral *p*-wave superconductors hosting Majorana fermions and $s^{+/-}$ superconductors believed to reside in a proximitized topological phase.

Topological crystalline insulators (TCIs): These are materials exhibiting topological order protected by spatial symmetries. An example is the recently proposed higher-order TIs (HOTIs), which are gapped on all faces but exhibit gapless corner and edge modes. Using our new techniques, we are investigating whether these states exist and whether interactions can gap out such topologically protected modes.

Strange metals: Interactions in these materials are so strong the scattering rate, $\tau^{-1} = k_B T/\hbar$, is believed to be "Planckian," i.e., determined by fundamental constants that set limits on the degree of quantum entanglement allowed in a many-body system. Using our quantum sensing techniques, we will search for hydrodynamic effects and identify the interactions that give rise to this bizarre state of matter.

Quantum Sensing and Quantum Materials (QSQM)	
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SLAC National Accelerator Laboratory	Thomas Devereaux, Benjamin Feldman, Kathryn Moler, Zhi-Xun Shen, Mariano Trigo
University of Illinois, Chicago	Dirk Morr

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Regenerative Energy-Efficient Manufacturing of Thermoset Polymeric Materials (REMAT)

EFRC Director: Nancy Sottos

Lead Institution: University of Illinois Urbana Champaign

Class: 2022 – 2026

***Mission Statement**: To advance the science of thermochemical reaction-diffusion processes in additive and morphogenic manufacturing and accelerate a transformative, circular strategy for thermoset polymeric and composite materials with programmed end-of-life.*

The Center for Regenerative Energy-Efficient Manufacturing of Thermoset Polymeric Materials (REMAT), a DOE BES Energy Frontier Research Center (EFRC) at the University of Illinois at Urbana-Champaign (UIUC) (lead) and its partner institutions: Sandia National Laboratories (SNL), Massachusetts Institute of Technology (MIT), Harvard University, Stanford University, and the University of Utah will address fundamental scientific challenges required to overcome barriers for energy efficient manufacturing of thermoset polymers and composites with realistic end-of-life strategies. Thermoset polymers and composites possess the necessary chemical and mechanical properties critical for achieving lightweight, durable structures in the energy, aerospace, and transportation industries, but the vast energy input required for initial manufacture (Gigajoules), long cure times to develop desired structural properties (hrs), and lack of end-of-life strategies render these materials unsustainable. The development of thermoset materials manufactured with a far lower energetic and environmental footprint is critically important to a carbon-neutral economy. The Center's goal is to discover thermoset resin formulations that enable (i) closed-loop controlled, energy-efficient additive manufacturing, (ii) moving beyond additive to nascent morphogenic manufacturing strategies, (iii) programmed end-of-life upcycling, and (iv) precise understanding of the chemistry and physics that control properties, performance, and multifunctionality for (re)use in structural materials.

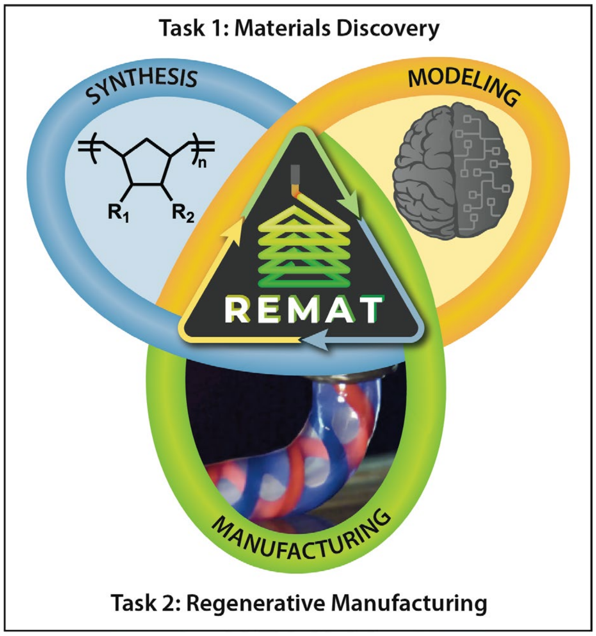

Fig. 1: Organization of the EFRC for Regenerative Energy-Efficient Manufacturing of Thermoset Polymeric Materials (REMAT).

The REMAT EFRC is addressing these multi-faceted scientific challenges through two highly collaborative and integrated Research Tasks (Fig. 1) that bring together synthesis, modeling, high throughput (HTP) experimentation, characterization, and machine learning (ML) to develop transformative manufacturing platforms that harnesses energy-efficient frontal polymerization. The Center will establish a new framework for fundamental research in frontal polymerization chemistry and manufacturing that develops state-of-the-art capabilities and expertise based on the realization that control and understanding of monomer chemical compositions are necessary for sustainable manufacture of thermoset structures. The research flow and subtasks are summarized in Fig. 2.

Task 1 – Materials Discovery for Circular Lifecycle will establish a new class of regenerative thermosetting polymers with controlled network evolution and generation-invariant properties. Subtask 1.1 focuses on resin chemistry. Cross-cutting Subtask 1.2 connects the input degrees of freedom (DOFs) with the output properties through the construction of statistical models. Subtask 1.3 uses automation to both prepare resins and make measurements of FP behavior.

Task 2 – Regenerative Manufacturing will combine additive manufacturing with a non-linear, dissipative curing process to achieve high energy efficiency. Resins developed in Subtask 1.3 are used by Task 2 for advanced manufacturing and materials properties assessment. Oligomers harvested from deconstructed thermosets returned to Task 1 for reactivation and reuse. At the intersection of Tasks 1 and 2 is the science of morphogenic manufacturing, which seeks to harness the mechanisms of dissipative structure formation to produce materials with exceptional durability.

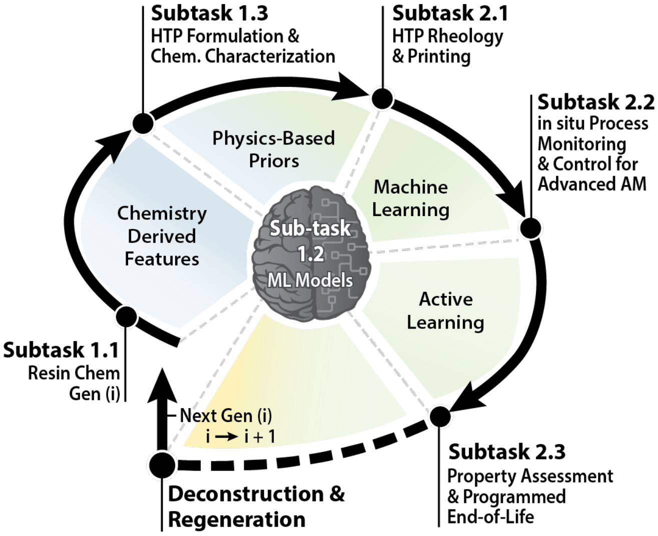

Fig. 2: Overview of end-to-end materials design and development cycle.

REMAT research will enhance the US economic competitiveness in industries ranging from lightweight electric vehicles to wind turbine blades. The integration of end-of-life strategies with materials discovery will enable multiple generations of reuse while reducing our dependence on petrochemical resources. The REMAT EFRC will also train the next generation of graduate students and postdoctoral researchers needed to carry out transformational research at the interface between sustainable materials chemistry and energy-efficient manufacturing in an innovative, inclusive, and interdisciplinary team-oriented environment.

Regenerative Energy-Efficient Manufacturing of Thermoset Polymeric Materials (REMAT)	
University of Illinois Urbana Champaign	Nancy Sottos (Director), Jeffrey Moore (Deputy Director), Jeff Baur, Randy Ewoldt, Philippe Geubelle, Sameh Tawfick
Harvard University	Jennifer Lewis
Massachusetts Institute of Technology	Rafael Gomez-Bombarelli, Jeremiah Johnson
Sandia National Laboratories	Leah Appelhans, Adam Cook
Stanford University	Yan Xia
University of Utah	Matthew Sigman

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Mechano-Chemical Understanding of Solid Ion Conductors (MUSIC)

EFRC Director: Jeff Sakamoto
Lead Institution: University of Michigan
Class: 2022 – 2026

Mission Statement*: to reveal, understand, model, and ultimately control the chemo-mechanical phenomena underlying the processing and electrochemical dynamics of ceramic ion conductors for clean energy systems.*

The recent emergence and discovery of new **ceramic ion conductors (CICs)** with fast ionic conductivity at near-ambient temperatures creates the opportunity to push the frontiers of electrochemical energy conversion and storage. The ability to replace traditional liquid or polymer electrolytes with ceramics has the disruptive potential to improve safety and enable next generation technologies including solid-state batteries with metal anodes, impermeable membranes to prevent crossover in redox flow batteries for long-duration energy storage (LDES), and intermediate temperature solid-oxide fuel cells and electrolyzers to propel the hydrogen economy. Enabling the next generation of electrochemical conversion and storage, however, requires fundamental research to understand and control the emergent **mechano-chemical** environments that arise when CIC materials are interfaced with other dissimilar materials. The overarching **scientific mission** of MUSIC is *to reveal, understand, and model, and ultimately control the chemo-mechanical phenomena underlying the processing and electrochemical dynamics of CICs for energy systems*.

This mission is supported by *specific hypotheses* that drive the research activities. To investigate and validate these hypotheses, MUSIC galvanizes a diverse team of internationally recognized leaders spanning the fields of electrochemistry, solid mechanics, ceramic synthesis and manufacturing, *in situ/operando* analysis, and multi-scale computational modeling. Within the MUSIC team, the convergence of the materials science, electrochemistry, solid mechanics, and manufacturing experts has the potential to solve critical problems that are central to CICs, yet would be challenging to solve by one discipline alone. Moreover, owing to growing industry, academic, and national lab workforce needs, MUSIC emphasizes career development through frequent and close interaction among early-career, mid-career researchers, and senior researchers, along with postdoctoral fellows, graduate, and undergraduate students.

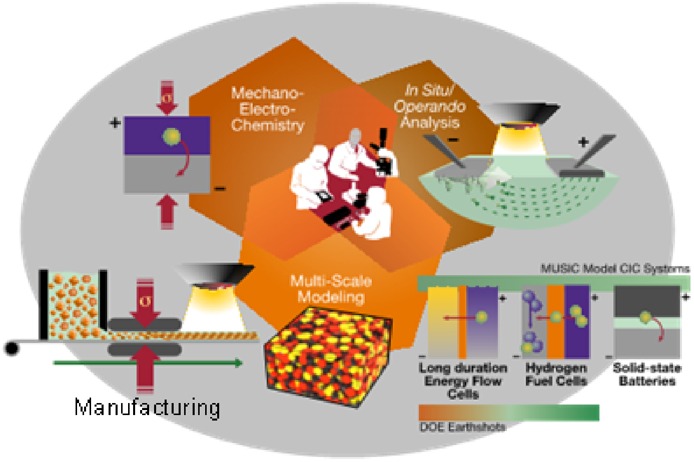

Figure 1. MUSIC will expand the field of mechano-electro-chemistry to accelerate progress toward DOE's Earthshots.

MUSIC has been created to achieve the overarching scientific mission detailed above and to meet the growing need for a concerted effort to integrate the fields of mechanics, chemistry, and electrochemistry to understand electro-chemo-mechanical phenomena underlying the synthesis and use of CICs for clean energy. The Senior Personnel in MUSIC have world-leading expertise in the areas needed to advance CIC science. (Figure 1) Connecting experiments to theory, leaders in the fields of multi-scale modeling with experience in mechano-electro-chemistry are integral to MUSIC. Augmenting the ability to better understand complex phenomena under dynamic conditions and at buried interfaces, MUSIC also includes key researchers that are advancing the state-of-the-art of *in situ*/*operando* and multi-scale modeling over all relevant length and time scales. To bolster efforts to create a viable and independent energy industry, processing and manufacturing science pervades across all themes within MUSIC. Most importantly, MUSIC acknowledges the role that postdocs and students will play in enabling science within the center and in future decades. MUSIC emphasizes training and fostering the next generation of scientists through robust bylaws, activities, promotion of workforce development, and continuous focus on supporting diversity, equity, and inclusion (DEI) efforts at all levels of the center.

Science Goals

The overarching Scientific Goals that will guide MUSIC are as follows:

SG1. How do mechanical stresses influence stability and charge-transfer kinetics of interfaces in CIC-based electrochemical systems as they evolve dynamically in time?
SG2. How are stress, electrochemistry, microstructure, and morphology related in CIC systems?
SG3. How do chemo-mechanical phenomena impact degradation pathways and at their interfaces?
SG4. How do chemo-mechanical phenomena in CICs influence the design and manufacturing of high-performance and resilient interfaces?

Research Goals

To achieve this ambitious vision, the four-year Research Goals of the MUSIC EFRC are as follows:

RG1. Develop *in situ/operando* platforms to reveal material dynamics at solid-solid, solid-liquid, and solid-gas interfaces and interphases of CICs spanning the atomic, meso, and microscales
RG2. Model and experimentally validate the coupling of chemical potential, stress state, and electric field in CICs to predict their influence on interfacial electrochemical kinetics and ionic transport
RG3. Reveal emergent chemo-mechanical degradation pathways across various length/time scales
RG4. Apply knowledge of the coupled mechanical and chemical phenomena in CICs to enable new manufacturing processes to improve resiliency, safety, and enable atomically precise control.

Mechano-Chemical Understanding of Solid Ion Conductors (MUSIC)	
University of Michigan	Jeff Sakamoto, Neil Dasgupta, Bart Bartlett, David Kwabi, Katsuyo Thornton, Michael Thouless
Massachusetts Institute of Technology	Yet-Ming Chiang, Bilge Yildiz
Northwestern University	Sossina Haile
University of Texas, Austin	David Mitlin, Donald Siegel
Georgia Institute of Technology	Mathew McDowell
Princeton University	Kelsey Hatzell
University of Illinois at Urbana-Champaign	Nicola Perry
Oak Ridge National Laboratory	Miaofang Chi
Purdue University	Partha Mukherjee

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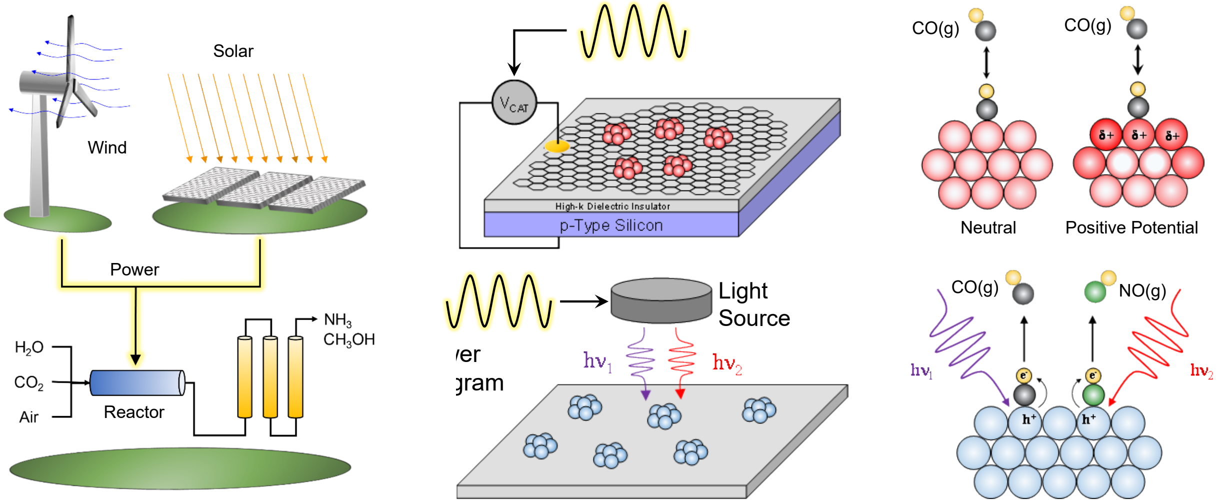
Center for Programmable Energy Catalysis (CPEC)
EFRC Director: Paul J. Dauenhauer
Lead Institution: University of Minnesota
Class: 2022 – 2026

Mission Statement*: To transform how catalysts control energy and accelerate chemistry.*

The emergence of low cost distributed power from wind turbines and solar photovoltaics provides attractive possibilities for using renewable electrons to drive thermocatalytic reactions in smaller, more efficient, and geographically-distributed catalytic reactors. To advance this goal, CPEC connects thermocatalysis to electrical power in the novel mechanism of providing power pulses to surface chemistry. The center's central thesis is that dynamic modulation of chemical bonding at catalytic active sites, achieved either by oscillating electron density or by photon-modified adsorbate-catalyst bonds, accelerates surface chemistry and provides control over reaction pathways to targeted products. Exciting results emerging in just the past few years have established the programmable concept and its promise for controlling surface chemistry. However, much remains to be discovered concerning programmable catalysis, its mechanisms, and fundamental behaviors. *The uniqueness of CPEC is its focus on fundamentals of complex forced dynamic surface kinetics* to understand the interplay between molecular-scale interactions, active site design, light and charge perturbations, and the dynamic changes in surface structure and adsorbate composition.



The center addresses the following integrative research objectives that advance fundamental understanding of programmable catalysis (i.e., catalysts perturbed with time).

- Understand charge distribution in catalyst active surfaces and sites under forced perturbation with light or charge.
- Construct reaction models that predict dynamic electronic perturbation of catalysts
- Understand programmable catalyst design and fabrication principles for high surface area and large active site density while retaining catalyst perturbation and dynamic enhancement
- Develop optimization strategies for predicting adsorbate/site/perturbation combinations *and* optimal oscillatory catalyst perturbations to accelerate and control energy-relevant chemistries

CPEC brings together expertise in heterogeneous catalysis, computational chemistry, and materials synthesis to create new catalytic devices that respond to light and charge. These two approaches to applying power locally to the surface have the commonality of oscillatory energy pathways of reacting molecules. Principles developed for one type of energy delivery mechanism apply to others. While the research addresses specific problems, general understanding of this new field of programmable catalyst will be developed in parallel to expand basic principles to a broader field of dynamic catalysts.

The center will establish new methods of operating catalysts dynamically via applied pulses of light with varying intensity, wavelength, and temporal application. Design of catalyst surfaces will be integrated with a focus on understanding the absorption of light and its effect on surface molecules and their reactions. This approach will identify new oscillatory conditions that efficiently use light to direct reaction pathways and accelerate reactions.

The center will also pursue fundamental understanding of a new thermocatalytic device called a 'catalytic condenser.' Comprised of an electrical insulator separating two conductive films, one of which is the catalyst layer, the condenser stabilizes charge (electrons or holes) at the catalyst surface, thereby altering the catalytic reaction. Research skills in materials synthesis and device design, computational chemistry, experimental kinetics, and reaction simulation will pursue an integrated approach to understanding the mechanisms of using charge to control chemistry, leading to faster and more controllable reactions beyond existing catalytic limits.

Center for Programmable Energy Catalysis (CPEC)	
University of Minnesota	Paul Dauenhauer (Director), Aditya Bhan, Matthew Neurock, C. Daniel Frisbie, Bharat Jalan
University of California Santa Barbara	Susannah Scott (Deputy Director), Phil Christopher Michael Gordon
University of Michigan	Eranda Nikolla
Clemson University	Rachel Getman
Massachusetts Institute of Technology	Yuriy Roman
University of Massachusetts Amherst	Omar Abdelrahman
University of Houston	Lars Grabow

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Center for Interacting Geo-processes in Mineral Carbon Storage (GMCS)

EFRC Director: Emmanuel Detournay

Lead Institution: University of Minnesota

Class: 2022 – 2026

Mission Statement*: To develop the fundamental science and engineering capability that will lead to realizing the full potential for the large-scale subsurface sequestration of CO2 via mineralization.*

A promising technology for permanently storing CO2 is sequestration via mineral carbonation in subsurface mafic and ultramafic rocks, e.g., basalt and peridotite, which are abundant in the Earth's crust. While this approach theoretically has the capacity to outpace anthropogenic CO2 emissions, its full potential is held back by the lack of knowledge of the processes that drive carbon mineralization.

Recent pilot scale projects, based on injecting dissolved CO2 in fractured basalt, have indicated that appropriate engineering can create conditions for effective carbon mineralization. The success of such an operation hinges on achieving a sustained dissolution-precipitation reaction between the host rock and dissolved CO2, whose rate is mainly influenced by the chemical potential of the host rock and the pH of the CO2 charge. The extent of the reaction throughout the reservoir is controlled by the ability of the CO2 charge to flow through the rock mass, along the fracture network and into the surrounding pore spaces. In turn, this flow is controlled by how the volume change, induced by the mineralization, clogs pores – retarding flow – or creates a network of cracks – promoting flow. Arriving at an effective operation for CO2 mineralization requires that these reaction, flow, and fracture processes, occurring across multiple space and time scales, work in a positive feedback loop. The objective of the Center is to arrive at a comprehensive understanding of the key interacting and multi-scale processes (Figure 1) that are necessary to achieve a successful carbonation operation.

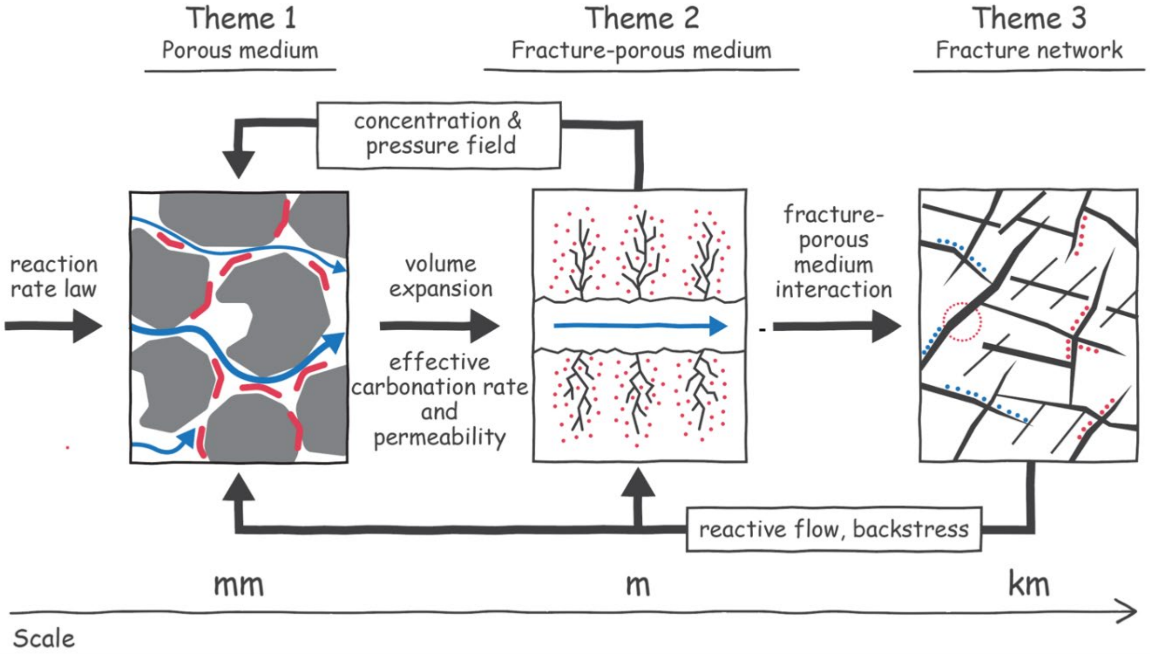

Figure 1. Interacting scales and processes in mineral carbon storage.

The Center will develop the mechanistic understanding of mineral carbonation in the subsurface by studying the reaction, flow, and fracture processes over three distinct scales (Figure 1):

(i) the porous medium scale (millimeter to decimeter) of the host rock, where the mineralization reactions and carbon storage takes place;
(ii) the fracture-porous medium scale (decimeter to meter), capturing an individual fracture from which the CO2 is delivered to the surrounding rock;
(iii) the fracture network scale (meter to kilometer) to describe how the CO2 charge is distributed within the reservoir by the natural or engineered fracture system.

Figure 1 illustrates the scales and processes of the theme domains together with their interconnections. The latter include research products and information that are passed not only from the small scales to larger scale but also from the large scales to smaller. Further note how each of these research themes is not restricted to a single knowledge domain. The threads of geomechanics, geochemistry, porous media transport, and sensing technology are tightly woven through each research theme. Knowledge about the key processes operating at each of these scales and the physics that bridge between the scales will be obtained through developing, adapting, and applying analytical and numerical modeling, physical experiments, and sensing techniques.

This intimate coupling and integration are necessary to provide answers to three fundamental overarching questions at the heart of the research efforts:

- *What are the key factors and processes that determine the evolving* CO_2 *mineralization rates (mass/time) in mafic and ultramafic rock masses?*
- *How do these factors and processes depend on the host rock lithology: mafic rocks (e.g., basalt) vs. ultramafic rocks (e.g., peridotite)?*
- *Once these factors and processes are identified and understood, how can the resulting models be deployed to generate hypotheses that can be tested at the scale of field operations?*

The successful outcome for sequestration of CO2 via mineralization will be measured by our ability to evaluate accurately the rate and amount of carbon that can be stored in a reservoir.

Center for Interacting Geo-processes in Mineral Carbon Storage (GMCS)	
University of Minnesota	Emmanuel Detournay, Bojan Guzina, Peter Kang, Joseph Labuz, Jia-Liang Le, Sonia Mogilevskaya, Vaughan Voller
Georgia Institute of Technology	Haiying Huang
Los Alamos National Laboratory	Bill Carey, Gowri Srinivasan, Hari Viswanathan
Northwestern University	Zdenek Bazant, Giuseppe Buscarnera, John Rudnicki
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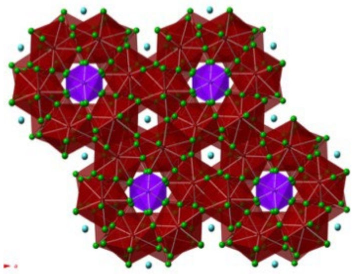
Center for Hierarchical Waste Form Materials (CHWM)

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.*

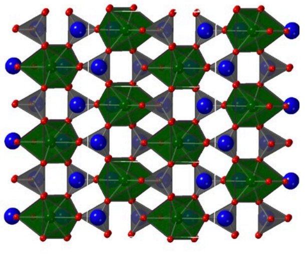
CHWM focuses on the exploration of hierarchical structures as potential waste form materials. The Center is focused on three general areas 1) radionuclide containing hierarchical materials, 2) materials to accommodate difficult to immobilize elements, and 3) understanding transport in multi-scale porous and hierarchical materials. Specifically, we are investigating 1) complex fluorides and oxides (phosphates, borates, silicates and germanates) accommodating Pu, Np, Am, and Cm; 2) tunnel structure materials, such as hollandites, designed to contain Cs and actinides; 3) MOFs with framework nodes consisting of and/or containing U, Th, Pu, and Np, with cavities to capture Tc and I; and 4) zeolite and geopolymer composites for sequestering Cs. We have successfully synthesized, characterized, and modeled numerous actinide, Tc and I containing phases, including $Na_xMPu_6F_{30}$, $Na_xMNp_6F_{30}$, and Cs_2MNpF_{16}; $(Ba,^{137}Cs)_{1.33}(Al/Cr,Ti)_8O_{16}$ hollandite; Th, U, and Pu MOFs; and Cs-containing geopolymer composites.

TRU Containing Oxides and Fluorides:



Structure of $Na_3AlPu_6F_{30}$

We are pursuing oxides and fluorides able to accommodate Pu(IV) and Np(IV) where we have synthesized and determined the structures of numerous new Np-containing fluorides of the type $Na_xMNp_6F_{30}$ (M = transition metals). In addition, we have extended this work to $Cs_2MNp_3F_{16}$ and will start to also target Pu(IV) containing analogs. New rare earth-containing double borate and phosphate phases were prepared, guided by our computing of stabilities to identify structures to readily accommodate Am(III) and Cm(III). This work resulted in the successful synthesis of single crystals of $K_3Am(PO_4)_2$, the first extended americium phosphate structure. This work is currently being extended to curium containing phosphates and borates. Single crystals of non-rad analogs belonging to the structural families discussed above were used in radiation effects studies and found to possess significant tolerance to radiation damage.



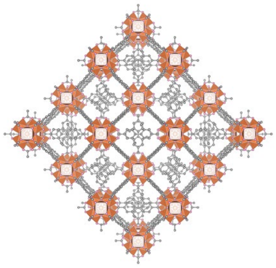
Structure of $K_3Am(PO_4)_2$

Tunnel Structure Materials:

Our research focuses on developing new hollandite compositions to sequester Cs. Using our ability to predict new tunnel structures, to determine their phase stability, as well as to develop detailed knowledge regarding their response to radiation, we prepared a series of Al- and Cr- substituted hollandites, $(Ba,Cs)_{1.33}(Al/Cr,Ti)_8O_{16}$, and determined their enthalpies of formation by high-temperature melt solution calorimetry. Interestingly, the results indicated enhanced thermodynamic stability with increased Cs content, which generally agreed with sublattice-based thermodynamic calculations. Structurally, the materials are complex and more detailed studies using neutron scattering measurements are underway. A parallel SEM/TEM study will correlate local structure features with macroscale determined stability of Cs. Atom probe tomography of these materials will be performed at the ORNL Nanophase Materials Science user facility. Irradiation studies reveal this class of materials have a high tolerance for radiation damage.

Novel Actinide-containing MOFs:

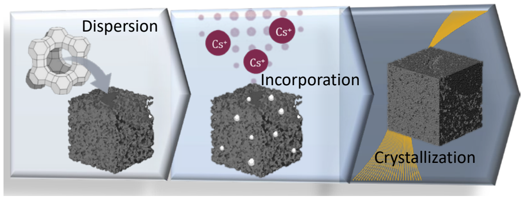
Actinide-MOFs (M = U, Th and Pu) were prepared via metal node expansion to create heterometallic nodes. Pu-MOFs have been prepared and structurally characterized. We experimentally demonstrated that the presence of capping linkers in certain Zr-MOFs significantly affects radionuclide release kinetics and to understand this process we developed a leaching model of radionuclides in Zr-MOF particles. Based on comparisons with actual experimental data, it is found that 1) the leaching of uranyl in Zr-MOF particles is a diffusion-controlled process; 2) the installation of capping linkers reduced the effective diffusion. The reduction was seen as proportional to the density of linkers, and dependent as well on the installation sequence of linkers; and 3) for Zr-MOF particles, the observed leaching kinetics can be explained by considering a thick surface layer where uranyl has an initial concentration gradient. The model correctly predicts the chemical diffusivity as a function of the installed linkers.



Single-crystal structure of Pu-MOF *Pu-Me$_2$BPDC-12*

Multi-Scale Porous Structures:

The Center has developed multi-scale and multi-phase materials that contain an amorphous phase (geopolymer) and crystalline particles (e.g., zeolite), and explored composites comprising geopolymers (amorphous alumino-silicates) and zeolites that are capable of entrapping and immobilizing radionuclides. We have explored different Faujasite zeolite compositions to determinate the thermodynamical stability of Na-Zeolite based structures. Using this information, we prepared (Na and Cs) zeolite/geopolymer composites and studied their composition, porosity, and crystallinity to explore their retention efficiency and performance as waste form. The crystallinity conversion of the geopolymer is being performed via hydrothermal synthesis to define the role of crystallinity in Cs trapping and immobilization. To further optimize the geopolymer compositions and composite microstructures to maximize the Cs ion exchange efficiency, a generic phase field model for a system containing four phases, i.e., zeolite, geopolymer, solution within the mesopores, and the solution, was proposed and we have developed the model and simulation code to capture the effect of anisotropic thermodynamic and kinetic properties, flow-field, and electric field on ion exchange kinetics.



Geopolymer composite for Cs uptake

Center for Hierarchical Waste Form Materials (CHWM)	
University of South Carolina	Hans-Conrad zur Loye (Director), Theodore Besmann (Deputy Director), Natalia Shustova
Alfred University	Scott Misture
University of Michigan	Gary Was, Kai Sun, Lumin Wang
Clemson University	Kyle Brinkman
University of Florida	Simon Phillpot
Savannah River National Laboratory	Jake Amoroso, David DiPrete
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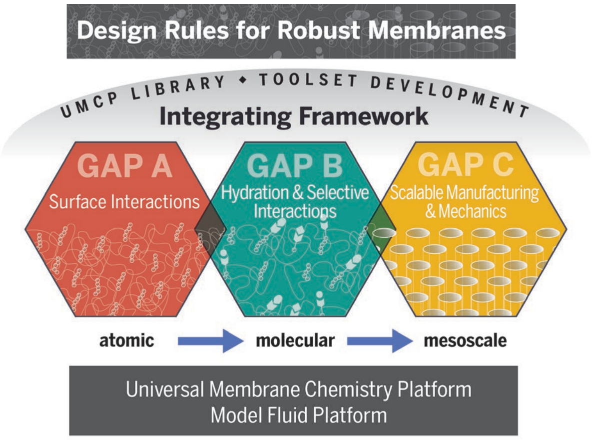
The Center for Materials for Water and Energy Systems (M-WET)

EFRC Director: Benny Freeman
Lead Institution: The University of Texas at Austin
Class: 2018 – 2026

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

Synthetic polymer membranes are widely used to purify water, mainly because they can be more energy efficient than competing (e.g., thermally based) technologies. However, water in energy applications is often heavily contaminated with a plethora of diverse organic and inorganic components. Current membranes were not designed (and are unsuitable) for such applications. Basic science knowledge gaps in thermodynamic and kinetic behavior of complex aqueous mixtures at interfaces, and the effect of such mixtures on the interfacial properties, limit our ability to translate fundamental understanding to transformative membrane materials design and manufacturing for energy/water applications. Moreover, current methods for synthesis and precision assembly of novel materials far from equilibrium do not allow for the scalable manufacturing of membranes with mesoscopic control over morphology for highly selective decontamination or resource recovery from such complex aqueous mixtures. The *Center for Materials for Water and Energy Systems* (M-WET) will fill these gaps in the understanding of fluids, materials, and non-equilibrium processing to catalyze the design of novel materials, highly selective solute/fluid interactions, mesoscopic structures, and transformative manufacturing strategies to prepare robust, high-performance membranes for energy applications.

Our central goal is to discover and design specific interactions with solutes of interest to create highly manufacturable, scalable, robust, selective, permeable membranes. Therefore, M-WET deploys **3 Gap Attack Platforms (GAPs)** to address fundamental knowledge gaps in water purification: *(A) Structure and Dynamics of Water and Solutes Near Interfaces*, *(B) Role of Hydration in Ion Transport and Separations*, and *(C) Fundamental Science of Membrane Manufacturing*. These **GAPs** work with an overarching ***Integrating Framework*** (**IF**) on *Bridging Between Systems/Developing the Bridging Toolset* to provide: (1) a common universal membrane chemistry platform (**UMCP**), (2) a model fluid platform (**MFP**), (3) high-throughput characterization methodologies to rapidly screen large regions of phase space, and (4) novel spectroscopic tools to provide unprecedented insight into water-solute-membrane interactions. A scalable, evolving library of **UMCP** materials will catalyze translation of discoveries between **GAPs** while the **MFP** provides continuity, coherence, and relevance



M-WET scientific framework, illustrating the proposed Gap Attack Platforms (GAPs) and Integrating Framework (IF) designed to uncover impacts of molecular/mesoscale water, solute, and polymer interactions on membrane properties. These GAPs are built on a foundation of a shared universal membrane chemistry platform (UMCP) and model fluid platform (MFP).

among research projects. Furthermore, we will develop/curate a highly functional, flexible database to organize M-WET's experimental data and provide a valuable information resource across M-WET and, ultimately, the scientific community. Via the **GAPs** and **IF**, M-WET will: (1) discover the key science drivers that link polymer structure and interfacial interactions to hydration, solute interactions, and fouling, (2) leverage fundamental understanding of ion solubility and transport in dry polymer systems to understand the role of hydration on water, ion, and solute transport in hydrated membrane materials relevant to water purification, (3) utilize high-throughput methods to rapidly identify selective interactants for incorporation into mechanically robust membranes, and (4) uncover the missing basic science that currently frustrates the facile manufacturing of high performance, robust, isoporous membranes via highly scalable non-equilibrium processes.

Today, specific interactions (and non-interactions) facilitate solute selectivity and fouling resistance, yet they cannot currently be "designed" *a priori* into membranes. To do so requires fundamental insights into the impact of functional groups on water dynamics near interfaces, an understanding of the impact of hydration on solute transport and membrane properties, and new routes to manufacture robust porous and nonporous membranes at scale. M-WET focuses on discovering and designing specific interactions with solutes of interest into highly manufacturable, scalable, robust, selective, permeable membranes. Motivated by the Basic Research Needs Report on Energy and Water, our specific goals are to: (1) design new interfaces with controlled topology and functionalities to achieve, for example, fouling-resistant surfaces and highly selective membranes; (2) precisely control mesoscopic material architecture to build novel, highly permeable, and selective membranes with rapid, component-resolved transport at multiple scales for resource recovery and producing fit-for-purpose water, while introducing design principles from (1); (3) develop novel imaging characterization tools for these systems; and (4) model multicomponent materials, fluid mixtures, and mesoporous architectures from atomistic to macroscale to radically transform membrane/materials systems' energy demands, resiliency, and efficiency.

The Center for Materials for Water and Energy Systems (M-WET)	
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University of California, Santa Barbara	Rachel Segalman (Associate Director), Christopher Bates, Phillip Christopher, Raphaële Clément, Glenn Fredrickson, Craig Hawker, M. Scott Shell, Todd Squires
Lawrence Berkeley National Labs	Ethan Crumlin, Dylan McReynolds, Gregory Su

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Multi-scale Fluid-Solid Interactions in Architected and Natural Materials (MUSE)
EFRC Director: Milind Deo
Lead Institution: University of Utah
Class: 2018 – 2024

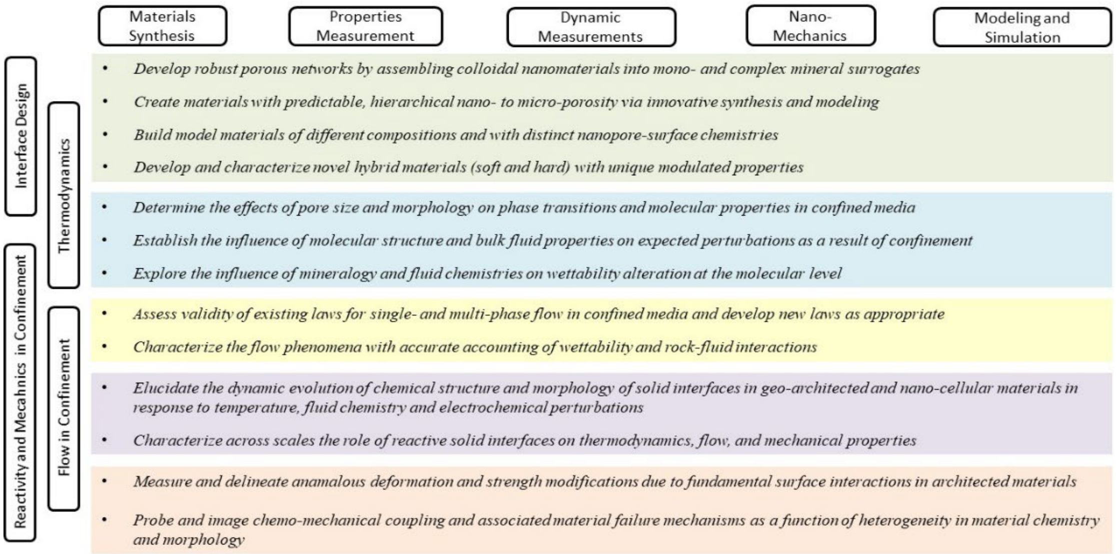
Mission Statement*: To synthesize geo-inspired materials with repeatable hierarchical heterogeneity and develop an understanding of transport and interfacial properties of fluids confined within these materials.*

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.



- *Develop robust porous networks by assembling colloidal nanomaterials into mono- and complex mineral surrogates*
- *Create materials with predictable, hierarchical nano- to micro-porosity via innovative synthesis and modeling*
- *Build model materials of different compositions and with distinct nanopore-surface chemistries*
- *Develop and characterize novel hybrid materials (soft and hard) with unique modulated properties*
- *Determine the effects of pore size and morphology on phase transitions and molecular properties in confined media*
- *Establish the influence of molecular structure and bulk fluid properties on expected perturbations as a result of confinement*
- *Explore the influence of mineralogy and fluid chemistries on wettability alteration at the molecular level*
- *Assess validity of existing laws for single- and multi-phase flow in confined media and develop new laws as appropriate*
- *Characterize the flow phenomena with accurate accounting of wettability and rock-fluid interactions*
- *Elucidate the dynamic evolution of chemical structure and morphology of solid interfaces in geo-architected and nano-cellular materials in response to temperature, fluid chemistry and electrochemical perturbations*
- *Characterize across scales the role of reactive solid interfaces on thermodynamics, flow, and mechanical properties*
- *Measure and delineate anamalous deformation and strength modifications due to fundamental surface interactions in architected materials*
- *Probe and image chemo-mechanical coupling and associated material failure mechanisms as a function of heterogeneity in material chemistry and morphology*

Figure 1. Design, Observations, Modeling and Simulations of Fluids in Architected Geo-Inspired Materials

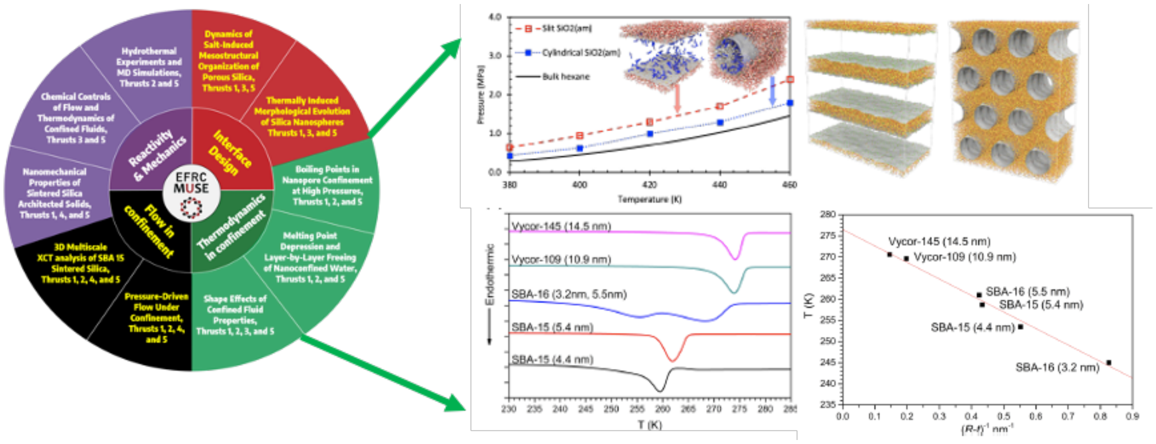


Figure 2. Experiments and Modeling Efforts span different thrusts, allowing rich collaborations.

Multi-scale Fluid-Solid Interactions in Architected and Natural Materials (MUSE)	
Idaho National Laboratory	Yidong Xia, Joshua Kane
The Pennsylvania State University	Adri van Duin (Thrust 4 Lead)
University of California, Davis	Subhash Risbud
University of Utah	Milind Deo (Director), Michael Bartl (Deputy Director), Darryl Butt, 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)
University of Wyoming	John Kaszuba

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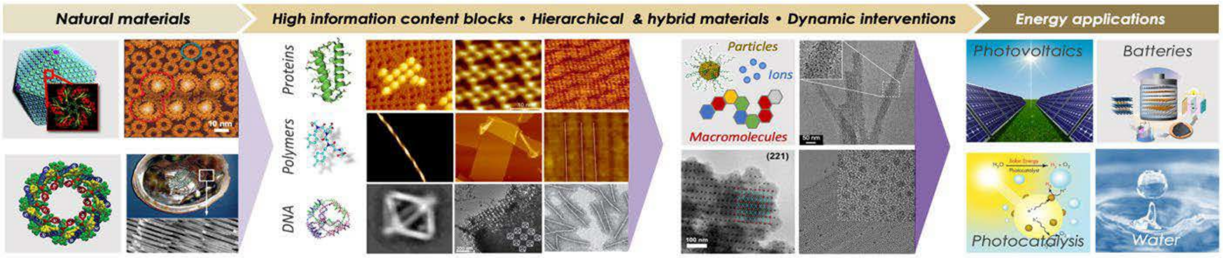
The Center for the Science of Synthesis Across Scales (CSSAS) EFRC Director: François Baneyx

Lead Institution: University of
Washington Class: 2018 – 2026

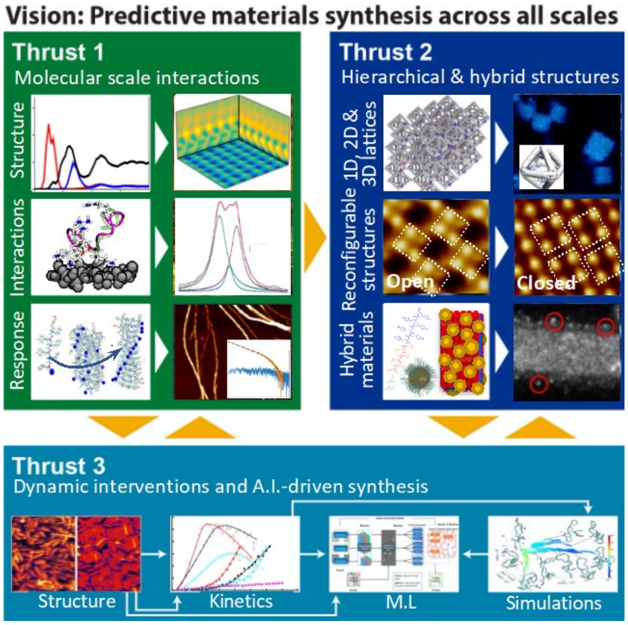
***Mission Statement**: Harness the complex functionality of hierarchical materials by mastering the design of high-information-content macromolecular building blocks that predictively self-assemble into responsive, reconfigurable, self-healing materials, and direct the formation and organization of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



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, the University of Tennessee Knoxville, the University of California San Diego and Columbia University, CSSAS will fill these knowledge gaps by tackling three research goals:



1. Determine the molecular-scale distribution and response of sidechains, solvent, and ions in the interfacial region of approaching building blocks, and atomic and nanoscale inorganic components targeted for assembly or directed nucleation. Understand how these distributions and responses give rise to the resulting interaction potentials that orchestrate materials formation at different scales.

2. Realize 2D and 3D hierarchical and hybrid materials by understanding how the interplay of interactions between disparate blocks, surfaces, solvent, and electrolytes determined in RG1 defines the energy landscapes across which hierarchy develops and inorganic nucleation proceeds.

3. Achieve adaptive control of synthetic outcomes and access non-equilibrium and metastable states of matter by integrating the tools of data science with *in situ* characterization and simulations, and by using external fields and localized changes in solution chemistry.

Our hypothesis-driven research starts by creating a set of systematically variable building blocks that span the scale of complexity – from large proteins to atomically- precise inorganic clusters. We accomplish the first goal by combining molecular-scale *in situ* observations with a hierarchy of simulation techniques that describe interactions and response dynamics. 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, we address 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 applied to responsive building blocks. In doing so CSSAS 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, Wendy Shaw
University of Chicago	Andrew Ferguson (Thrust 1 Lead)
University of California San Diego	Akif Tezcan
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GRAND CHALLENGES INDEX

TRANSFORMATIVE OPPORTUNITIES INDEX

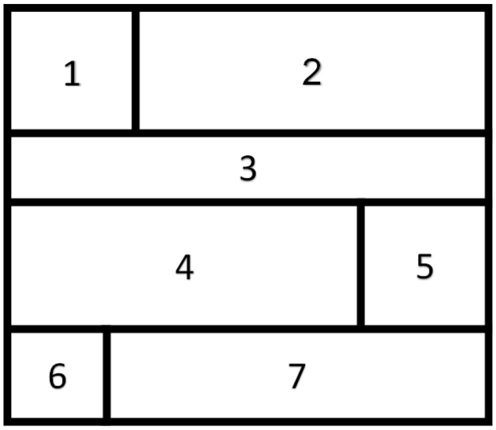
BES REPORTS INDEX

ADDENDUM

FRONT COVER IMAGE

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

1. Center for High Precision Patterning Science (CHiPPS) led by Patrick Naulleau at Lawrence Berkeley National Laboratory (pg. 27)
2. Institute for Cooperative Upcycling of Plastics (iCOUP) led by Aaron Sadow at Ames Laboratory (pg. 3)
3. Institute for Quantum Matter (IQM) led by Collin Broholm at Johns Hopkins University (pg. 23)
4. Center for Quantum Sensing and Quantum Materials (QSQM) led by Peter Abbamonte at University of Illinois at Urbana-Champaign (pg. 85)
5. Center for Novel Pathways to Quantum Coherence in Materials (NPQC) led by Joel Moore at Lawrence Berkeley National Laboratory (pg. 25)
6. Center for Materials for Water and Energy Systems (M-WET) led by Benny Freeman at University of Texas at Austin (pg. 97)
7. Bioinspired Light-Escalated Chemistry (BioLEC) led by Gregory Scholes at Princeton University (pg. 55)



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