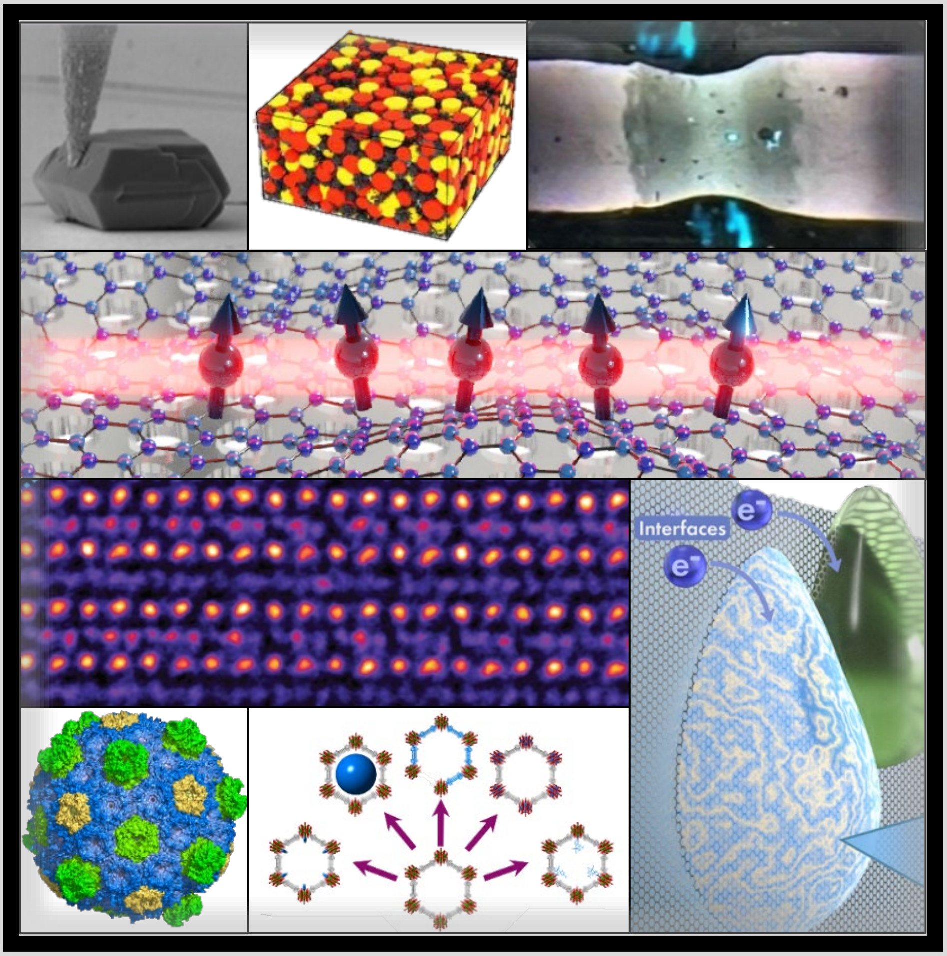
Energy Frontier Research Centers

Technical Summaries



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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 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 the scientific community in 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 107 centers across 43 states and the District of Columbia. Biennial funding opportunities for 4-year awards started in 2014. The 2024 funding opportunity marked the 7th class of these Energy Frontier Research Centers awards, with a targeted competition in advanced manufacturing, including polymers and co-design of materials and processes to revolutionize fabrication science for microelectronics and quantum information science, and environmental management of nuclear waste tanks. These 10 centers will join the 34 continuing EFRCs that focus on key fundamental energy science areas including advanced manufacturing, energy storage, environmental management, hydrogen, microelectronics, nuclear, quantum information science, separations, solar, and the subsurface to make 44 active Energy Frontier Research Centers. Each center has its own unique mission that supports the Department of Energy's initiatives in advancing basic energy research across the United States.

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Ordered by Institution and then by Director

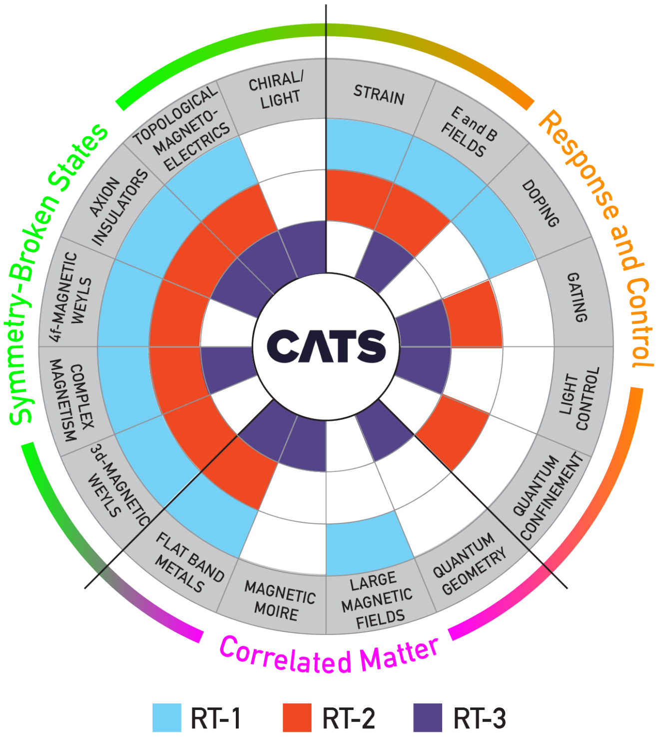
Center for the Advancement of Topological Semimetals (CATS)
EFRC Director: Robert McQueeney
Lead Institution: Ames Laboratory
Class: 2018– 2026

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

The intersection between topology, magnetism, and electronic correlations is an exciting scientific frontier 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) is achieving its mission through a highly collaborative fundamental research program, with the vision to provide the innovation required to realize the promises of this exciting frontier.

CATS groundbreaking successes in the discovery, synthesis, theory, and manipulation of 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 are expanding this frontier to encompass distinctive spin and charge symmetry-broken ground states in topological materials.



CATS integration. CATS goals are in the outer circle and the principal phenomena and capabilities are color-coded by Thrust.

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 well-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 ability to control their phenomena, which, in turn, challenges and advances our scientific understanding. CATS is developing 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 layered assemblies of topological materials with special emphasis on nonlinear optical and transport responses. These thrusts 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.

Center for the Advancement of Topological Semimetals (CATS)	
Ames Laboratory	Robert McQueeney (Director), Paul Canfield, Adam Kaminski, Ben Ueland, Linlin Wang
Argonne National Laboratory	Anand Bhattacharya (Deputy Director), Ivar Martin (Deputy Director)
Boston College	Qiong Ma
Harvard University	Suyang Xu, Ashvin Vishwanath
Los Alamos National Laboratory	Ross McDonald (RT-1 leader), Johanna Palmstrom
Massachusetts Institute of Technology	Joe Checkelsky, Pablo Jarillo-Herrero (RT-3 leader)
University of California – Santa Barbara	Susanne Stemmer (RT-2 leader)
University of Illinois – Chicago	Thomas Searles
University of Waterloo	Anton Burkov

Contact: Robert McQueeney, Director, mcqueeney@ameslab.gov
(515)-294-7712, https://cats.ameslab.gov

Institute for Cooperative Upcycling of Plastics (iCOUP)
EFRC Director: Aaron Sadow
Lead Institution: Ames National Laboratory
Class: 2020 – 2028

Mission Statement*: To establish the molecular and macromolecular scientific principles governing deconstruction and reconstruction of polymers that enable sustainable manufacturing from energy-rich plastics.*

The iCOUP team is investigating the selective catalytic conversions of hydrocarbon polymers into valuable chemicals and new recyclable materials to establish new transformative manufacturing science principles that enable a circular economy for plastics. Plastics are essential in the global economy, as reflected by production of new polymers surpassing 400 million tons in 2023, for applications that touch all aspects of modern life. Many of the advantages of plastics are diminished by environmental impacts of their initial manufacture and end-of-life. Manufacturing of plastics consumed the equivalent of 6–8% of the crude oil and natural gas produced worldwide. Approximately 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. The 'upcycling' conversion of used polyolefins into new chemicals with defined end-of-life, including new feedstocks for chemically recyclable polymers, could avoid the negative impacts of the conventional plastic economy. 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.

In general, selective conversions of macromolecules require catalytic sites to react with polymers on length and time scales suited to molecules and they require that the architecture supporting the catalytic site also influences the polymer chains' behavior on macromolecular length and time scales. Nature's approaches to selective biopolymer conversions, including DNA synthesis and repair as well as cellulose deconstruction, achieve precision using enzymes that operate under the constraints of these multiscale phenomena. Inspired by nature, we are creating catalytic materials and chemically recyclable polymers that address molecular and macromolecular length scales to enable circular transformative manufacturing. Our approaches involve processive mechanisms in which the catalyst creates a template to influence the conformations of adsorbed polymer chains (**Fig 1A-B**) to break polymer backbone linkages or install new reactive groups at specific positions. We will use these functionalized species, both mesomonomers and macromolecules, to create chemically recyclable polymers with high-quality mechanical properties (**Fig 1C**).

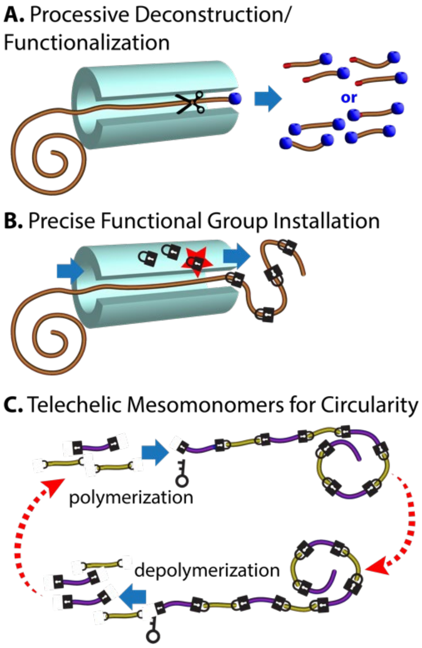


Fig. 1. Three approaches for selective deconstruction or manufacture of plastic materials.

We will study how materials with mesoscale architectures govern conversions of long polymer chains, while adapting catalytic principles from molecular chemistry to access desired functionality, through the following Objectives:

(1) Invent catalytic reactions for breaking and functionalizing C–C and C–H bonds to transform polymers into upcycled mesomolecular intermediates.
(2) Install catalytic active sites for C–C functionalization within hierarchical support architectures to produce uniformly functionalized, value-added chemicals from discarded polymers.

The iCOUP team will adapt these new transformations for the synthesis of telechelic mesomonomers and recyclable polyolefin-like materials, while establishing the new chemical and materials science principles that provide recyclable plastic materials with desirable mechanical properties.

(3) Synthesize telechelic mesomonomers from discarded plastics, leveraging length-selective cleavage and functionalization to install reactive groups at both chain ends.
(4) Reassemble these telechelic mesomonomers into recyclable-by-design polymers and establish their structure-thermomechanical property relationships.

These transformations, as well as the mechanical properties of the resulting plastics, are driven by intermolecular interactions and size-dependent dynamics of populations of hydrocarbon polymer chains (**Fig 2**). We will study these phenomena to establish the mechanisms that control polymer upcycling reactions and re-polymerizations.

(5) Design unique mechanistic motifs that drive selectivity across length and time scales.
(6) Measure polymer-surface interactions and dynamics to provide quantitative multiscale thermochemical properties and kinetics underpinning polymer conversions.

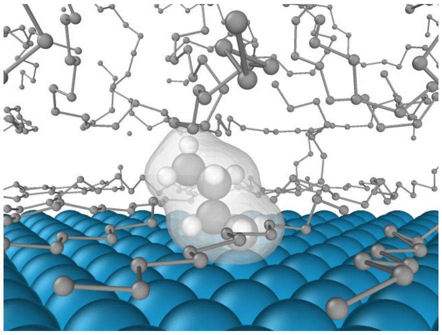


Figure 2. Model of long and short chains reacting on metal surfaces.

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, repolymerized, 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.

Institute for Cooperative Upcycling of Plastics (iCOUP)	
Ames Laboratory	Aaron Sadow (Director), Frédéric Perras, Wenyu Huang
Argonne National Laboratory	Massimiliano Delferro (Deputy), Byeongdu Lee
Cornell University	Geoffrey Coates, Anne LaPointe
Northwestern University	Erik Luijten, Kenneth Poeppelmeier
University of California, Santa Barbara	Susannah Scott, Mahdi Abu-Omar
University of Illinois at Urbana-Champaign	Baron Peters
University of South Carolina	Andreas Heyden
University of Southern California	Megan Fieser
University of Michigan	Anne McNeil
University of Florida	Russell Bowers

Contact: Aaron Sadow, Director, sadow@iastate.edu
515-294-8069, https://www.ameslab.gov/institute-for-cooperative-upcycling-of-plastics-icoup

Advanced Materials for Energy-Water Systems (AMEWS)

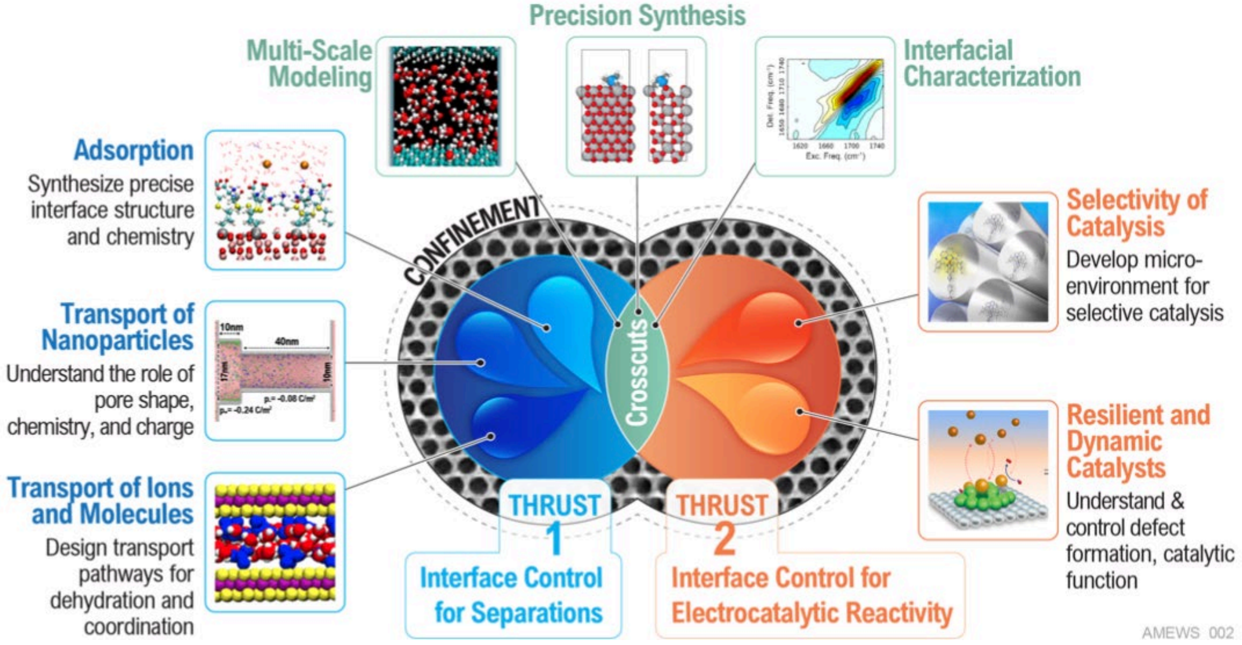
EFRC Director: Seth Darling
Lead Institution: Argonne National Laboratory
Class: 2018 – 2026

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.

Advanced Materials for Energy-Water Systems (AMEWS)	
Argonne National Laboratory	Seth Darling (Director), Juan de Pablo*, Jeffrey Elam, Paul Fenter, Giulia Galli* (Deputy Director), Alex Martinson (Deputy Director), Karen Mulfort, Paul Nealey*, Pietro Papa Lopes, Matthew Tirrell*
University of Chicago	Chibueze Amanchukwu, Nicholas Lewis, Chong Liu, Dmitri Talapin, Andrei Tokmakoff (* - joint appointee)
Northwestern University	George Schatz, Chris Wolverton
Princeton University	Kelsey Hatzell

Contact: Seth Darling, Director, darling@anl.gov
630-252-4580, https://www.anl.gov/amews

Ultra Materials for a Resilient, Smart Electricity Grid (ULTRA)

EFRC Director: Robert J. Nemanich
Lead Institution: Arizona State University
Class: 2020 – 2026

Mission Statement*: To understand fundamental phenomena in UWBG materials – including synthesis, defect and impurity incorporation, electronic structure at interfaces, the interaction of electrons and phonons at high fields to achieve extreme electrical properties, and phonon phenomena that affect thermal transport. The Future Grid Co-Design Ecosystem enables communication across all levels of the science and technology.*

A resilient, smart electricity grid is necessary to integrate multiple energy sources, power storage capabilities and diverse electrical needs; ultra-wide bandgap (UWBG) semiconductors have been identified as a crucial enabling materials technology. These UWBG semiconductor and dielectric materials (or 'Ultra materials') present a new realm for high field transport, electron-phonon interactions, and heat transport. Understanding these materials and their novel properties will enable efficient energy conversion and control and a great reduction in size where a substation could be replaced by a suitcase sized power converter (Fig. 1).

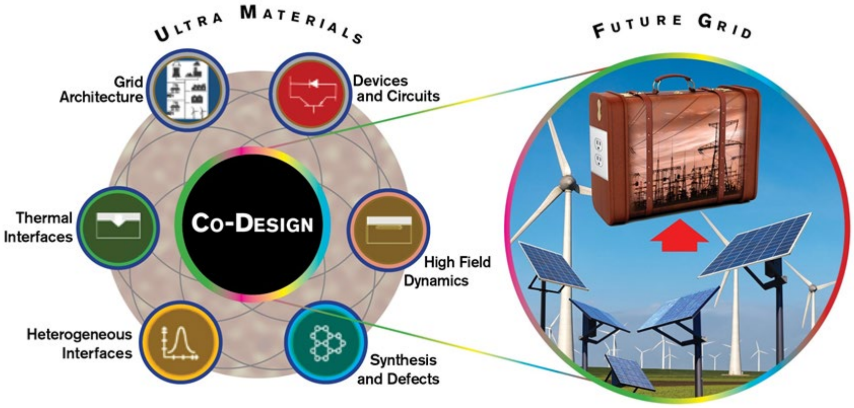


Figure 1. Schematic of ULTRA EFRC Thrusts as part of a Co-design ecosystem that will impact the future electricity grid including the "Substation in a Suitcase" concept.

ULTRA EFRC is focused on basic science challenges in four thrust areas: (1) growth, defects and impurities, (2) heterogeneous interfaces, (3) carrier dynamics and high field transport, (4) thermal energy transport and interfaces. The UWBG semiconductors of interest include cubic diamond, hexagonal AlN and $B_xAl_{1-x}N$ alloys, and oxide, nitride and fluoride materials as dielectrics and alternative semiconductors. ULTRA EFRC established the Future Grid Co-design Ecosystem and has developed a knowledge-base of UWBG materials and properties to "Reinvent the Electricity Grid." The research will establish a roadmap to achieve a breakdown field greater than 10 MV/cm, current densities greater than 100,000 A/cm^2, interface-constrained effective heat dissipation > 1 kW/cm^2, and voltage (current) slew rates of 250 V/ns (100 A/ns). Significantly, ULTRA is providing critical input into design simulation tools for a new generation of high-power devices and power conversion modules and continues to work with grid architect researchers to emphasize the most relevant properties of the UWBG semiconductors as part of the Future Grid Co-design Ecosystem.

The goals of ULTRA EFRC are:

Synthesis, Defects and Impurities: Understand growth chemistry, defect formation and impurity incorporation during scalable synthesis of UWBG materials, substitutional doping and co-doping to span intrinsic to degenerate doping, distributed polarization doping of polar UWBG semiconductors, and *in situ*, real time characterization for machine learning and precision design.

Heterogeneous Interfaces: Efficiently employ different growth systems (MBE, MOCVD, PECVD, ALD) to enable epitaxial UWBG heterostructures, and dielectric heterostructures. Develop interface control to impact band offsets, interface states, polarization design and doping, Fermi level control and charge transfer - modulation doping.

Carrier Dynamics and High Field Transport: Employ designed field control structures for high field carrier transport, breakdown phenomena and electron-phonon interactions at UWBG heterostructures and dielectric interfaces. Consider multi-mode transport for high current including photoexcitation to describe breakdown in UWBG Materials.

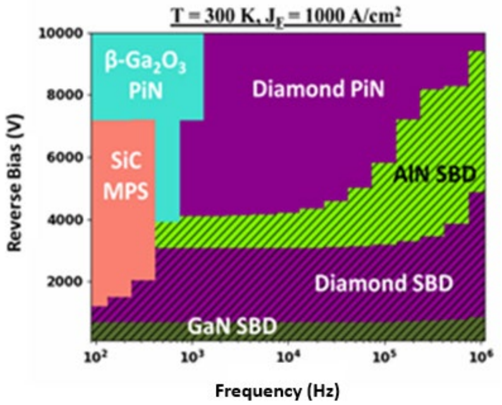


Figure 2. Color map showing a comparison of different material and device technologies based on the lowest power dissipation at a given operating point in terms of reverse bias and frequency for a fixed current density.

Thermal Energy Transport and Interfaces: Understand thermal transport in UWBG materials, the dependence on doping, isotope and composition, and at their interfaces, including effects from phonon dispersion and scattering, electron-phonon interactions and the electrothermal behavior of materials. Determine how interface disorder and interfacial layers affect thermal transport across boundaries.

Future Grid Co-design Ecosystem: Integrate theory, synthesis, fabrication, and characterization in the four research thrusts as a co-design process to rapidly meet the research goals and objectives of ULTRA EFRC. Utilize ULTRA research outcomes to make predictive assessments of their potential impact in power electronics as shown in Fig. 2, and the electric grid network, and identify critical areas for UWBG research.

Ultra Materials for a Resilient, Smart Electricity Grid (ULTRA)	
Arizona State University	Robert Nemanich (Director), Stephen Goodnick (Deputy Director), Nidhin Kurian Kalarickal, Fernando Ponce, Marco Saraniti, Arunima Singh, David Smith
University of Alabama-Birmingham	Mary Ellen Zvanut
University of California – Los Angeles	Alexander Balandin
University of California – Riverside	Richard Wilson
Cornell University	Debdeep Jena, H. Grace Xing
Michigan State University	Timothy Grotjohn
Morgan State University	Michael Spencer
Ohio State University	Hongping Zhao
Rice University	Yuji Zhao
Sandia National Laboratories	Jack Flicker, Robert Kaplar
Stanford University	Srabanti Chowdhury (Science Collaboration Director)
University of Bristol, UK	Martin Kuball

Contact: Robert J. Nemanich, Director, robert.nemanich@asu.edu
(480) 965-2240, https://ultracenter.asu.edu/

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.

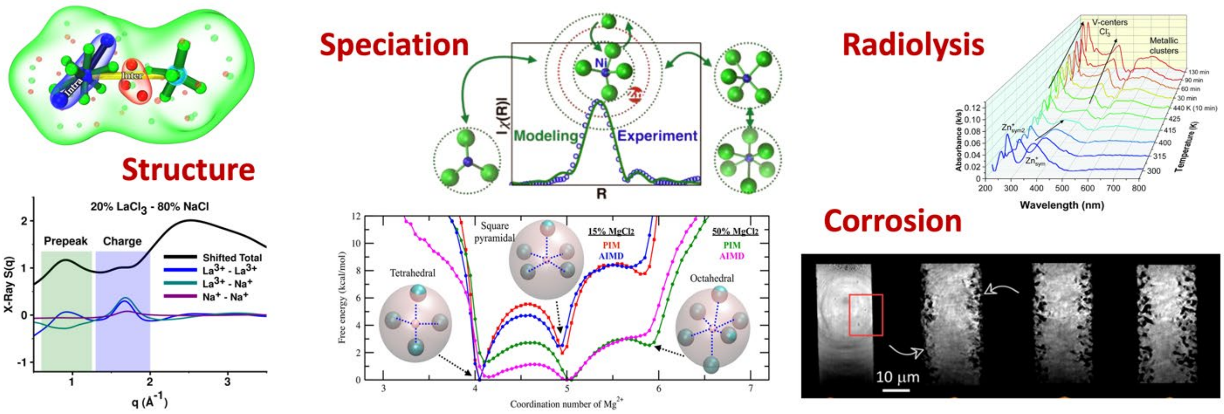
Though made from simple ionic components, molten salts are complex, structured liquids that are subject to intricate physical and chemical processes, particularly in the extreme environments encountered in reactors. Their behavior with solutes, including fuel actinides, and in contact with reactor materials, 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.



Molten Salts in Extreme Environments (MSEE)	
Brookhaven National Laboratory	James Wishart (Director), Simerjeet Gill, Anatoly Frenkel, Benjamin Ocko
Oak Ridge National Laboratory	Shannon Mahurin (Deputy Director), Vyacheslav Bryantsev, Sheng Dai, Alexander Ivanov, Santanu Roy
Idaho National Laboratory	Ruchi Gakhar (Thrust 2 Leader), Gregory Holmbeck (Radiation Crosscut Leader), Kaustubh Bawane, Jacy Conrad
University of Iowa	Claudio Margulis (Thrust 1 Leader)
Stony Brook University	Yu-Chen Karen Chen-Wiegart (Thrust 3 Leader)
University of Michigan	Katsuyo Thornton
University of Notre Dame	Jay LaVerne, Edward Maginn
University of Tennessee	Sheng Dai (JA with ORNL)
University of Wisconsin	Adrien Couet

Contact: James Wishart, Director, wishart@bnl.gov
631-344-4327, https://www.bnl.gov/moltensalts/

Breakthrough Electrolytes for Energy Storage and Systems (BEES2)

EFRC Director: Burcu Gurkan

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

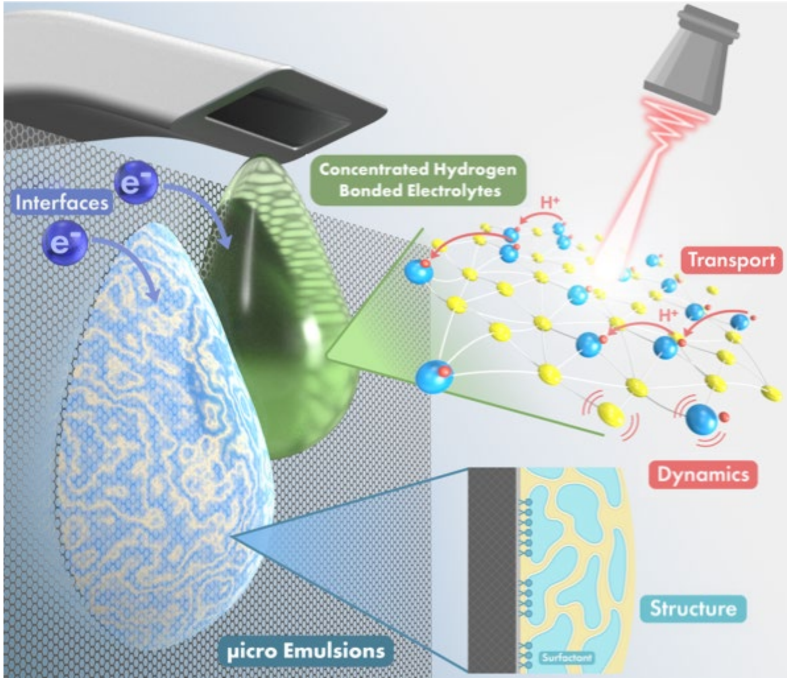


FIGURE 1. Electrolytes of interest to BEES2 EFRC are represented along with bulk (H-bonding network dynamics, proton conduction, structure, *etc.*) and interfacial (electron transfer, redox reactions, *etc.*) properties of importance.

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.

Breakthrough Electrolytes for Energy Storage and Systems (BEES2)	
Case Western Reserve University	Burcu Gurkan (Director), Robert Savinell (Chief Scientist), Rohan Akolkar, Clemens Burda, Kasandra Nagy (Program Manager), Jesse Wainright (Thrust 2 Co-Lead), Robert Warburton
Hunter College at CUNY	Steve Greenbaum
New York University	Mark Tuckerman
Ohio State University	Josh Sangoro (Thrust 1 Co-Lead)
Texas A&M University	Emily Pentzer (Thrust 1 Co-Lead), David Powers
University of Illinois Urbana-Champaign	Joaquin Rodriguez-Lopez
University of Notre Dame	Edward Maginn
University of Tennessee Knoxville	Mark Dadmun, Manolis Doxastakis, Thomas Zawodzinski (Thrust 2 Co-Lead)

Contact: Burcu Gurkan, burcu.gurkan@case.edu, 216-368-4182,
https://engineering.case.edu/research/centers/breakthrough-electrolytes-for-energy-storage

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 performance, 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, the AI modeling and inverse design research thrust (blue circle) will develop new AI models, including physics-informed neural networks (PINN) and multiscale deep

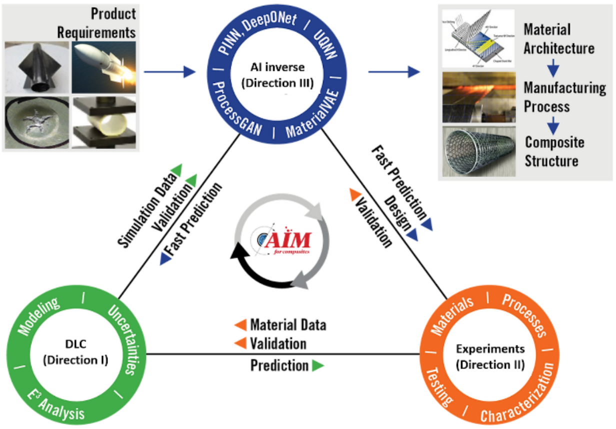


Figure 1. *Project overview: iterative loop among the DLC, AI modeling, and Experiments research thrusts, then AI models enable inverse design workflow.*

neural operators (DeepONet), to efficiently map 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 University of Delaware's Center for Composite Materials, the University of Florida's Center for Manufacturing Innovation, 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). 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. To further demonstrate and validate the DLC and AI models, we have recently incorporated testbeds (injection molding, thermoforming, and 3D printing) capable of fabricating varied geometries. Thus, the optimal composite material and its manufacturing process are 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 capture the material behavior at multiple length scales (impurities, complex compositions), the effects of manufacturing processes (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 boundaries 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 make 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	Gang Li (Associate Director), Zhen Li, Qiong Zhang, Mik Carbajales-Dale, Feng Luo, James Sternberg
University of Delaware / Clemson	Srikanth Pilla (Director)
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, Ivan Radev
Savannah River National Laboratory	Dale Hitchcock
Pacific Northwest National Laboratory	Kevin Simmons (RT-3 Leader), Yao Qiao. David Mayberry, Daniel Merkel, Khaled Shahwan, Maria Swita

Contact: Srikanth Pilla, Director, spilla@udel.edu
302-831-3182, https://www.aimforcomposites.com/

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

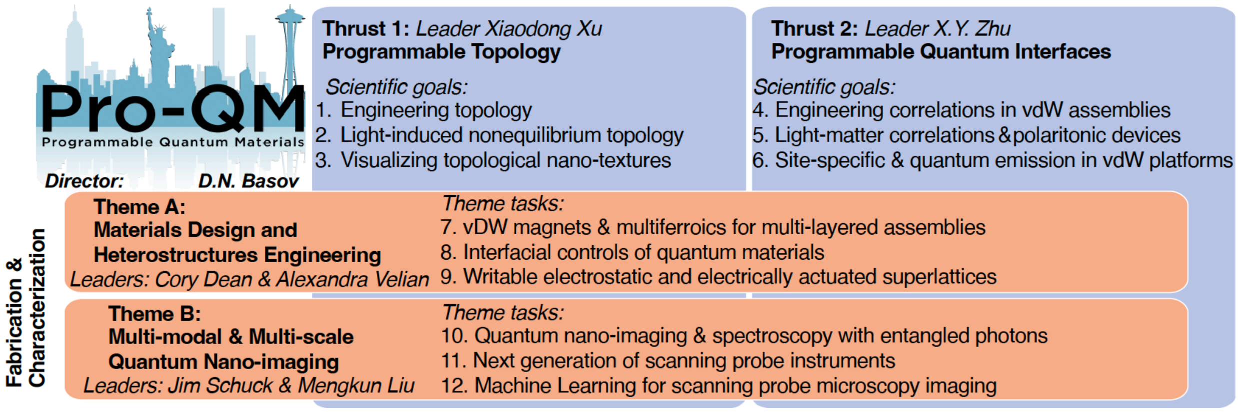
The scientific goals of the four-year program of Pro-QM are organized in two interdependent Research Thrusts (Fig. 1.1). Thrust 1 will create and image interacting topological states in van der Waals (vdW) quantum materials with emphasis on dynamic manipulation of band topology and on-demand topological 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.

Since the inception of the award, the Pro-QM team has published 89 collaborative works, including numerous articles in *Nature* and *Science*. Approximately 30% these papers acknowledge EFRC funding as the primary source of support. Many of these publications are highly collaborative, with up to nine co-authors. The Pro-QM team is therefore solidly on track toward meeting and surpassing its research targets pertaining to the mission of quantum phases on demand.



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

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

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

Theme B:
Multi-modal & Multi-scale Quantum Nano-imaging
Leaders: Jim Schuck & Mengkun Liu
Theme tasks:
10. Quantum nano-imaging & spectroscopy with entangled photons
11. Next generation of scanning probe instruments
12. Machine Learning for scanning probe microscopy imaging

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

Programmable Quantum Materials (Pro-QM)	
Columbia University	Dmitri N. Basov (Director), Ana Asenjo-Garcia Cory R. Dean, Milan Delor, James Hone, Michal Lipson, James McIver, Andrew J. Millis, Abhay Pasupathy, Raquel Queiroz, Xavier Roy, P. James Shuck, Xiaoyang Zhu
University of Washington	Jiun-Haw Chu, David H. Cobden, Alexandra Velian, Di Xiao, Xiaodong Xu, Matt Yankowitz
Brookhaven National Laboratory	Valentina Bisogni, Mengkun Liu

Contact: D.N.Basov, Pro-QM EFRC Director, db3056@columbia.edu
212-853-1320, https://qunatum-materials.columbia.edu

Center for Alkaline Based Energy Solutions (CABES)
EFRC Director: Héctor D. Abruña
Lead Institution: Cornell University
Class: 2018 – 2026

Mission Statement*: 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. We seek a detailed understanding of the nature, structure, and dynamics of fuel cell systems operating in alkaline media. 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): **SD-1**. What factors govern electrocatalysis in alkaline media? **SD-2.** How do we understand and control transport in alkaline media? **SD-3.** What makes energy materials durable in alkaline media?

The center's integrated approach is 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 as well as experimental and computational tools necessary for, and critical to, a fundamental understanding of these processes. We foresee the center as providing the basis for ushering an alkaline-based energy technology society with emphasis on fuel cells and electrolyzers.

CABES addresses fundamental issues of critical need and importance in energy conversion technologies, guided by the challenges articulated in the relevant DOE reports, Basic Research Needs and in support of two Energy Earthshots (Hydrogen and Long Duration Storage). CABES' advances are highlighted in authoritative and comprehensive review articles (Y. Yang, C. R. Peltier…H. D. Abruña; "*Electrocatalysis in Alkaline Media and Alkaline Membrane-Based Energy Technologies*" Chem. Rev. 2022, 122, 6, 6117–6321). Qihao Li, Andrés Molina Villarino… David A. Muller, Geoffrey W. Coates, Piotr Zelenay, and Héctor D. Abruña "*Anion Exchange Membrane Water Electrolysis: The Future of Green Hydrogen*" Journal of Physical Chemistry C (2023), 127(17), 7901-7912

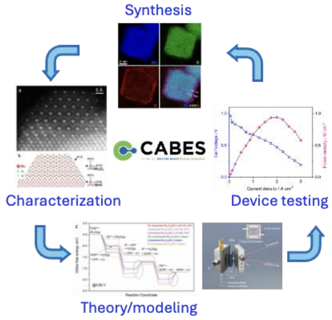


Fig. 1. SD1 synergistic research loop illustrating interactions between/among the different components of the team.

CABES' SD-based approach revolves around synergistic loops enabled by the center's three cross-cutting disciplines: materials synthesis and characterization, modeling/data science and *operando* analysis (including device testing). Fig. 1 presents the loop for SD-1. Work of each discipline within the loop informs the others; i.e. theory may suggest new catalysts to synthesize, or characterization helps refine modeling. We have expanded our previously successful strategy of strain-engineering the surfaces of nanoparticles by tuning the core composition to enhance reactivity. 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, as well as strain. 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.2)

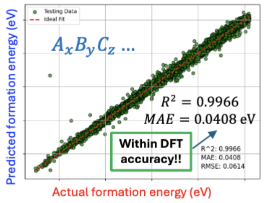


Fig.2 Predicted and actual formation energies of multicomponent catalyst candidates.

Science Driver 2 is focused on how the architecture, especially of electrolyte/membrane materials, and interfaces within them, influences ion, molecular, and electronic transport in electrochemical systems under alkaline conditions. The relationships between architecture and transport are investigated through the same integrated approach employed in SD-1. Systematic synthesis alkaline exchange membranes (AEMs), correlated ionomers, (including new synthetic strategies that afford structural control) and the respective interfaces between/among ionomers, catalysts, and supports are analyzed with analytical and theoretical/computational local probes of critical parameters. We also assess the transport properties that result from these structures, through measurements of water uptake, water permeability, hydroxide ion conductivity, in AEM fuel cells and AEM water electrolyzers.

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 high performing and durable electrolytes be designed and synthesized. We are also focused on understanding interactions of membranes and ionomers with catalysts/supports.

Theory and Computation, as a cross-cut effort, informs the most promising directions to pursue by employing Cahn-Hilliard dynamical studies, Joint Density Functional Theory modeling and machine learning potentials. Fig. 2. Additionally, machine learning and artificial intelligence approaches are accelerating the search for new electrocatalysts and improving understanding of transport phenomena.

CABES *operando* methods cross-cut effort, has developed unique experimental capabilities including cryo-TEM for the study of membranes and ionomers in the electrode environment. We developed a liquid cell incorporating electrodes on the viewing membrane in a transmission electron microscope to observe electrochemical processes in their native environment at the nanometer scale. We have acquired an electron microscope exclusively dedicated to operando measurements (Perseus), which we believe is the only one of its kind. (Fig.3) CABES has developed X-ray nano-diffraction and nano-fluorescence analysis to quantify structural defects in catalysts for OER. We have developed a new tool for localizing, with nanometer resolution, regions with high strain, by leveraging nanoprobe diffraction, and will extend this method to nanoimaging to enable us to examine the impact of strain and defects on electrocatalysis in alkaline media. With unique characterization tools to examine the structure and strain distributions in catalyst nanoparticles at atomic resolution, and at the nanoscale, *in-situ*, *operando* and device testing of both components and membrane electrode assemblies, we have information to guide the synthesis, and test and inform the modeling.

Fig.3 Electron microscope dedicated to operando studies.

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

Contact: Paul Mutolo, Executive Director, pfm2@cornell.edu
607-255-4928, https://cabes.cornell.edu/

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 objective 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 are leveraging 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 focus will be achieved by combining new syntheses, *in situ/operando* characterization, molecular modeling, and machine learning approaches applied to catalysts, sorbents, and membranes use in clean energy technologies such as H_2 generation and CO_2 capture and conversion.

We are leveraging 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) direct air capture of CO_2; (ii) green generation of H_2 via water splitting / electrolysis; and (iii) electrochemical conversion of clean feedstocks (such as H_2 and CO_2) into useful products. 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) Synthesize a series of new adsorbents, (electro)catalysts, or membranes that meet or exceed established benchmarks in terms of cost, performance, and stability.
2) Create a library of the impact of emerging contaminants on adsorbent, (electro)catalyst, and membrane structure and activity.
3) Develop computational models to guide the rational design of new materials and predict their stability in realistic contaminant environments.

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 modeling of emerging contaminant interactions with functional solids for targeted materials design. An overview of the various research tracks for Phase III is shown in Figure 1.

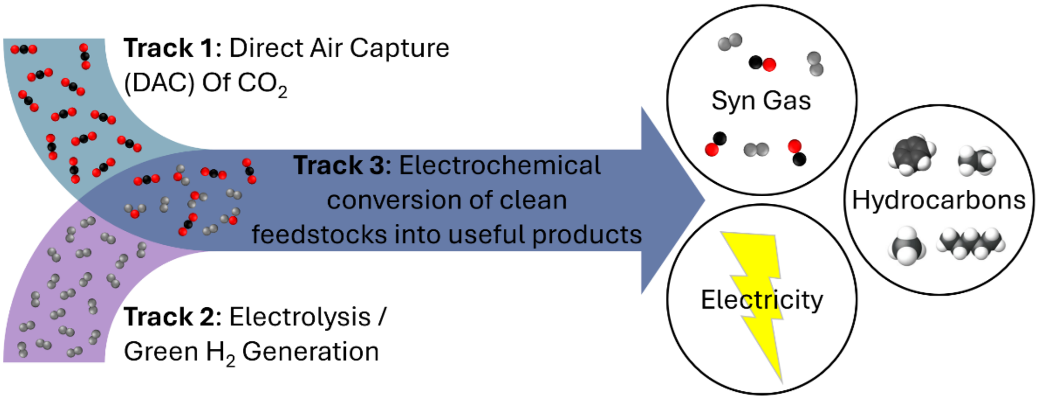


Figure 1. Schematic overview depicting the interrelated clean energy technology Tracks which are the focus of UNCAGE-ME.

Teams comprised of senior personnel are grouped into Research Thrusts based on expertise, and members from each Thrust will address basic scientific questions associated with the technologies described in each of the three Tracks. The three thrusts are: (i) New Materials Synthesis; (ii) Realistic Contaminant Effects; and (iii) Computation & Informatics. The New Materials Synthesis Thrust focuses on designing, synthesizing, and testing new functional materials with tailored properties for H_2 generation and CO_2 capture, as well as their electrochemical conversion into other useful products. The Realistic Contaminant Effects Thrust will focus on advanced *in situ/operando* structural and performance analysis of the new materials for H_2/CO_2 processing in realistic process streams. 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 Computation & 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 new materials discovery.

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

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

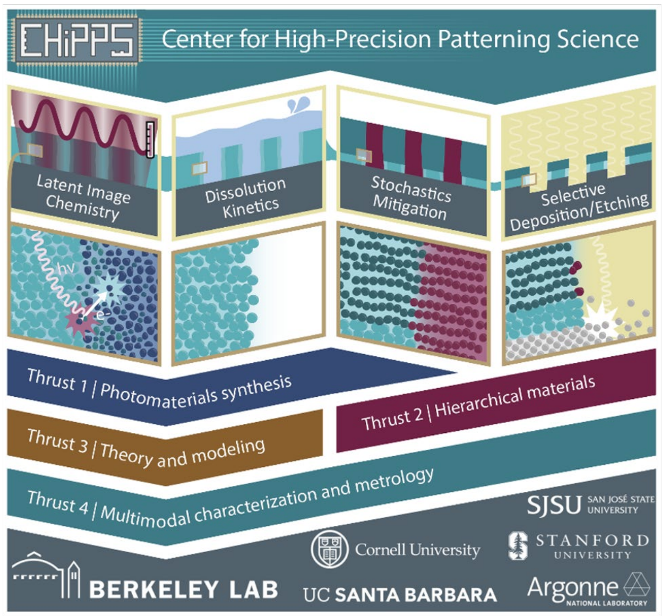
Center for Thermal Transport under Irradiation (TETI)	
Idaho National Laboratory	David Hurley (Director), Boopathy Kombaiah, Krzysztof Gofryk, Zilong Hua, Amey Khanolkar, Kaustubh Bawane, Chao Jiang, Linu Malakkal, Brelon May, Shuxiang Zhou
Oak Ridge National Laboratory	Michael Manley
Airforce Research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

Contact: David Hurley, Director, david.hurley@inl.gov
(208) 360 6374, https://teti.inl.gov

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 $526B/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 pursues 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 with precise architectures 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 further integrates 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 are 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 develops fundamental understanding of the chemical reactions involved in pattern formation. It informs Thrust 1 by implementing quantitative theory and modeling for rational design of patterning materials and performs experimental validation with Thrust 4.
4. **Thrust 4. Multimodal Characterization.**
   The cross-cutting Thrust 4 develops 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)	
Lawrence Berkeley National Laboratory	Samuel Blau, Weilun Chao, Brett Helms, Frances Houle, Oleg Kostko, Ricardo Ruiz (Director), Bruno La Fontaine (Deputy Director), Cheng Wang
Argonne National Laboratory	Paul Nealey
Cornell University	Christopher Ober
San Jose State University	Dahyun Oh
Stanford University	Stacey Bent
University of California, Santa Barbara	Rachel Segalman

Contact: Ricardo Ruiz, Director, ricardo.ruiz@lbl.gov
510.495.2893, https://chipps.lbl.gov

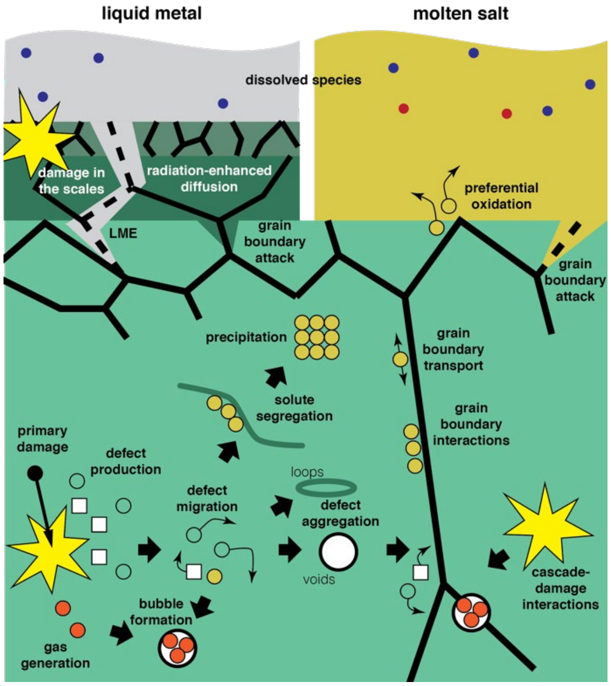
Fundamental Understanding of Transport Under Reactor Extremes (FUTURE)

EFRC Director: Blas Pedro Uberuaga
Lead Institution: Los Alamos National Laboratory
Class: 2018 – 2026

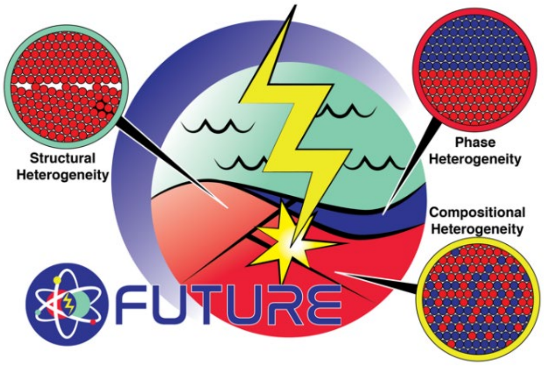
Mission Statement*: To understand how the coupled extremes of irradiation and corrosion work in synergy to modify the evolution of materials by coupling experiments and modeling that target fundamental mechanisms.*

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)	
Los Alamos National Laboratory	Laurent Capolungo, Edward Holby, Aaron Kohnert, Nan Li, Blas Uberuaga (Director), Yongqiang Wang
University of California, Berkeley	Mark Asta, Peter Hosemann (Deputy Director), Andrew Minor, Raluca Scarlat
Pacific Northwest National Laboratory	Tiffany Kaspar, Dan Schreiber, Sandra Taylor, Kayla Yano
Arizona State University	Farida Selim
North Carolina State University	Djamel Kaoumi
University of Texas, San Antonio	Elizabeth Sooby
University of Virginia	John Scully

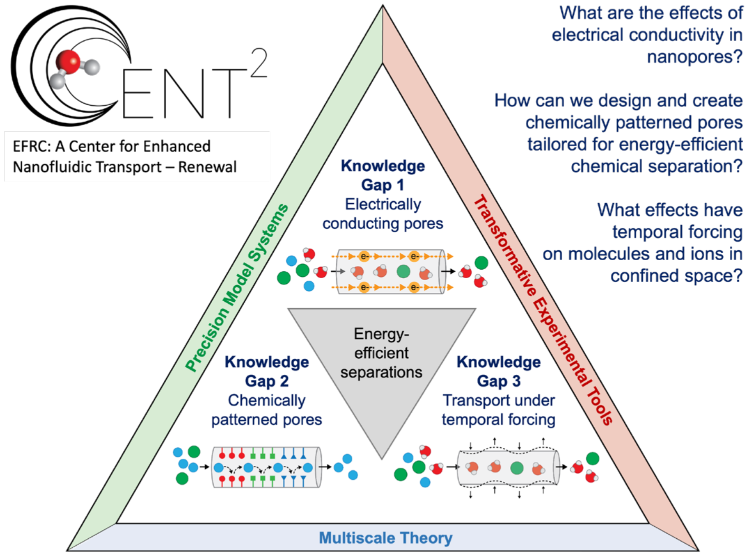
Contact: Blas Pedro Uberuaga, Director, blas@lanl.gov
505-667-9105, https://m.lanl.gov/future

Center for Enhanced Nanofluidic Transport Phase 2 (CENT^2)
EFRC Director: Michael S. Strano
Lead Institution: Massachusetts Institute of Technology
Class: 2018 – 2026

Mission Statement*: To address 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 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



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

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)	
Massachusetts Institute of Technology	Michael Strano (Director), Martin Bazant, Daniel Blankschtein, Heather Kulik
University of Maryland	YuHuang Wang (Deputy Director), John Cumings, John Fourkas
Lawrence Livermore National Laboratory	Aleksandr Noy, Tuan Anh Pham
University of California Irvine	Zuzanna Siwy, Javier Sanchez-Yamagishi
Yale University	Menachem Elimelech, Amir Haji-Akbari
Stanford University	Arun Majumdar
University of Texas, Austin	Narayana Aluru
University of Florida	Charles Martin

Contact: Michael Strano, Director, strano@mit.edu
617-324-4323, https://cent.mit.edu

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). 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.

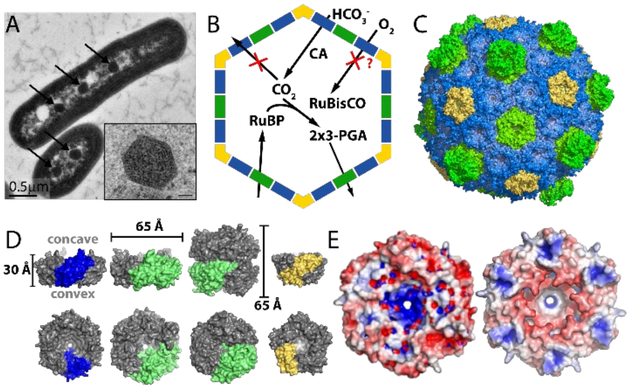
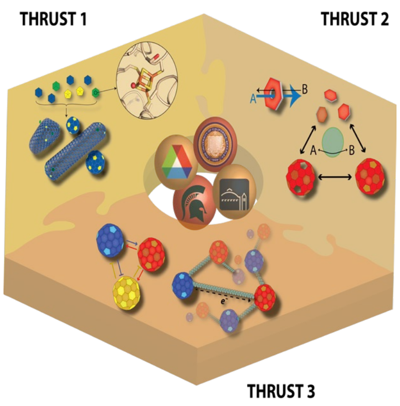


Figure 1. Structure, function, and context of BMCs. A) Cyanobacteria with carboxysomes (arrows); inset, close up view of a 200 nm carboxysome. B) Schematic of carboxysome function. C) Structure of a 6.5 MDa BMC shell determined by a combination of X-ray crystallography and Cryo-EM[3]. D) Basic building 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 in energy-converting chemistries; Thrust 3 investigates the mesoscale assembly and organization of compartmentalized catalysis.

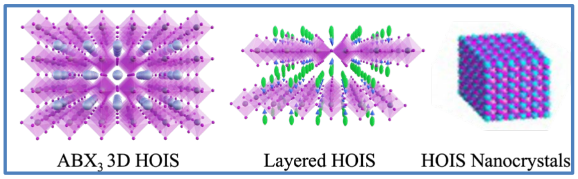
Center for Catalysis in Biomimetic Confinement (CCBC)	
Michigan State University	Cheryl Kerfeld, Eric Hegg, Michael Feig, Robert Hausinger, Kristin Parent, Markus Sutter, Michaela TerAvest, Josh Vermaas
Argonne National Laboratory	David Tiede, Karen Mulfort, Oleg Poluektov, Lisa Utschig, Xiaobing Zuo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

Contact: Damien Sheppard, Operations Manager, sheppa@msu.edu
https://www.ccbc-efrc.org/

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.

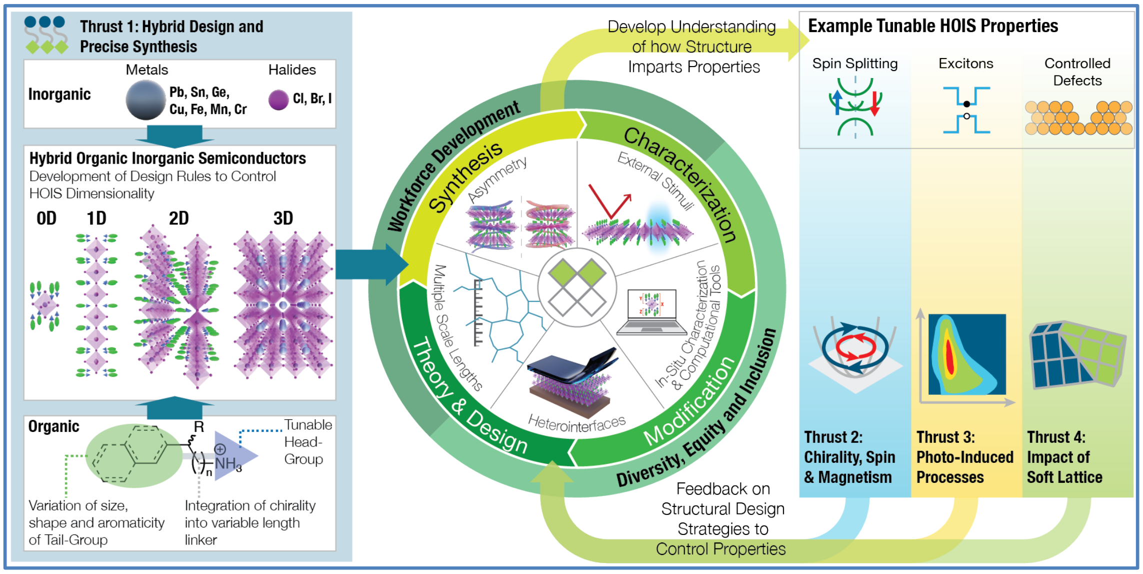


HOIS are unconventional semiconductors with properties that vary continuously from those associated with organic semiconductors having tightly bound Frenkel-type excitons characterized by large exciton binding energies, to those associated with inorganic semiconductors exhibiting delocalized Wannier-type 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)	
Duke University	David Mitzi, Volker Blum, Adrienne Stiff-Roberts
National Renewable Energy Laboratory	Matthew C Beard, Joseph Luther, Kai Zhu, Jeffrey Blackburn, Peter Sercel
North Carolina State	Dali Sun
University of Wisconsin, Madison	Yuan Ping
University of Utah	Valy Vardeny
University of North Carolina, Chapel Hill	Jinsong Huang
University of Toledo	Yanfa Yan
University of Colorado Boulder	Mike Toney, Joseph Berry, Seth Marder
San Diego State University	Yong Yan

Contact: Matthew C. Beard, Director, choise.efrc@nrel.gov
303-384-6781, www.choise-efrc.org

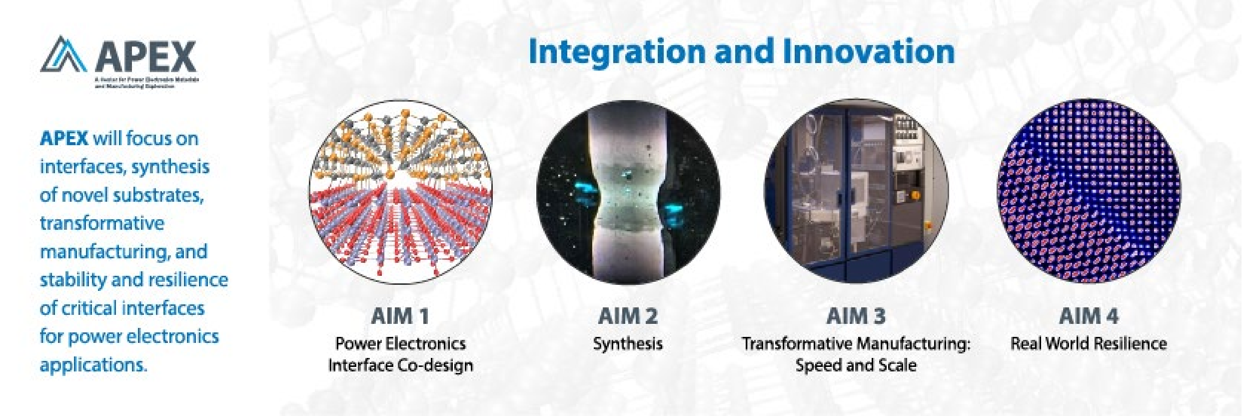
A Center for Power Electronics Materials and Manufacturing Exploration (APEX)

EFRC Director: Nancy M. Haegel
Lead Institution: NREL
Class: 2024 – 2028

Mission Statement*: To expand interdependent materials and manufacturing choices for substrates, ultrawide-bandgap semiconductors, contacts, thermal sinks, and critical interfacial layers and advance fundamental understanding of structurally, chemically, thermally, and electrically dissimilar interfaces.*

Next-generation power conversion technologies have the potential to significantly improve energy efficiency and reduce global energy consumption. **APEX** (A Center for Power Electronics Materials and Manufacturing Exploration) will enable a wider selection of materials and manufacturing methods built for speed and scale for next-generation power electronics by understanding and elucidating chemical, physical, thermal, and electrical requirements at dissimilar interfaces. We are motivated by a co-design vision for “A on B,” where “A on B” can encompass a range of materials and interfaces that promise to unlock new functionality but may be more difficult to achieve from a synthesis standpoint. The APEX team, an interdisciplinary community of scientists, is driven by the greatest energy challenge of our time: the urgent need to innovate our way to a highly electrified, sustainable, and clean energy ecosystem.

Transformative device design and associated manufacturing of power electronics will be required to fully achieve broad electrification of transportation, industrial process decarbonization, and the development of the “grid of the future.” APEX combines theory-driven interface co-design, synthesis, scalable manufacturing techniques, and advanced characterization to address the needs for better thermal management and increased power capacity, while also reducing device size and weight and increasing real-world resilience.

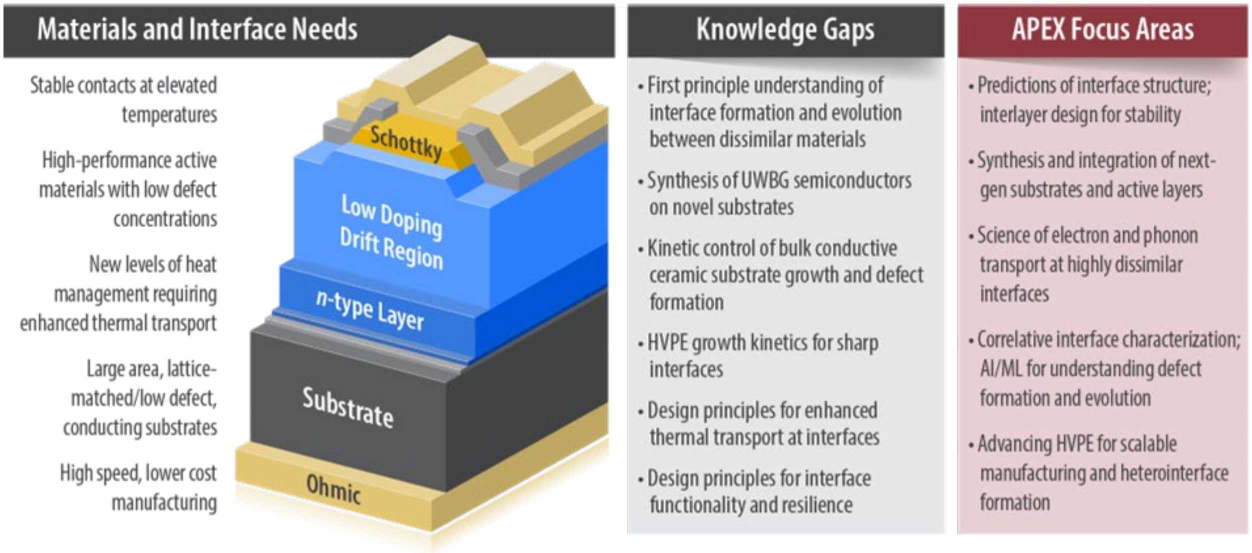


APEX will focus on interfaces, synthesis of novel substrates, transformative manufacturing, and stability and resilience of critical interfaces for power electronics applications.

Selection of the best materials for a given function is often limited by the challenges of connecting highly dissimilar materials in stable, reliable, and resilient ways. APEX will expand the range of interdependent materials choices. Our focus will be borides, nitrides, carbides, and/or oxides, which hold potential to advance performance and transform manufacturing approaches for power electronics by enabling the design of smaller devices that can handle more current. These materials include semimetallic borides and metallic carbides, which are ideal partners for advancing AlGaN technology, with favorable lattice and thermal expansion coefficients, as well as novel oxide interfacial layers applicable to Ga_2O_3 and SiC. APEX will conduct basic research in theory-guided interface design for stabilization and thermal conductance, novel substrates, kinetics of hydride vapor phase epitaxy (HVPE) growth, and interface resilience, using a co-design approach to enable demanding combinations of electronic, thermal, and mechanical properties that are required to boost power electronics performance.

The desire to move from lateral to vertical architectures to reduce electrical resistance and manage current crowding issues means that the substrate (whether dictated by growth or by liftoff and integration) will participate in future device operation in a more critical way. Today's "tyranny of the substrate" (constraints related to the synthesis, doping and cost of suitable large, high-quality crystals) limits both the substrate materials available for power electronics and the quality of the active layers grown on them. This research aims to lift those constraints.

The aims of APEX are: (1) develop design rules for broader heterointegration through a co-design approach that includes first-principles theory and experimental studies of formation and design of highly dissimilar interfaces; (2) demonstrate novel growth of conductive bulk single crystal carbide and boride substrates and related innovation in active layer heterogeneous integration, liftoff, and transfer; (3) transform manufacturing via HVPE to achieve both scale and speed, including studying AlGaN growth kinetics, incorporating in-situ diagnostics, and combining HVPE with layer liftoff as a manufacturing pathway; and (4) understand and control interfacial phases, reactions, and evolution to further the science for real-world performance, stability, and resilience, applying multi-modal, multi-scale structural, thermal, and electronic characterization to study interface stability under relevant temperatures and electric fields and using new machine learning/artificial intelligence (ML/AI) tools to increase correlative insight.



A Center for Power Electronics Materials and Manufacturing (APEX)	
National Renewable Energy Laboratory (NREL)	Nancy Haegel (Director), Kirstin Alberi, Stephan Lany, John Simon, Steven Spurgeon, Brooks Tellekamp, Andriy Zakutayev
Morgan State University	Michael Spencer, MVS Chandrashekhar
Johns Hopkins University	Tyrel McQueen
Colorado School of Mines	Vladan Stevanovic
University of Virginia	Patrick Hopkins
Argonne National Laboratory	Jessica McChesney
Kyma Technologies	Heather Splawn, Jacob Leach

Contact: Nancy M. Haegel, Director, Nancy.Haegel@nrel.gov
303-384-6548

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.

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.

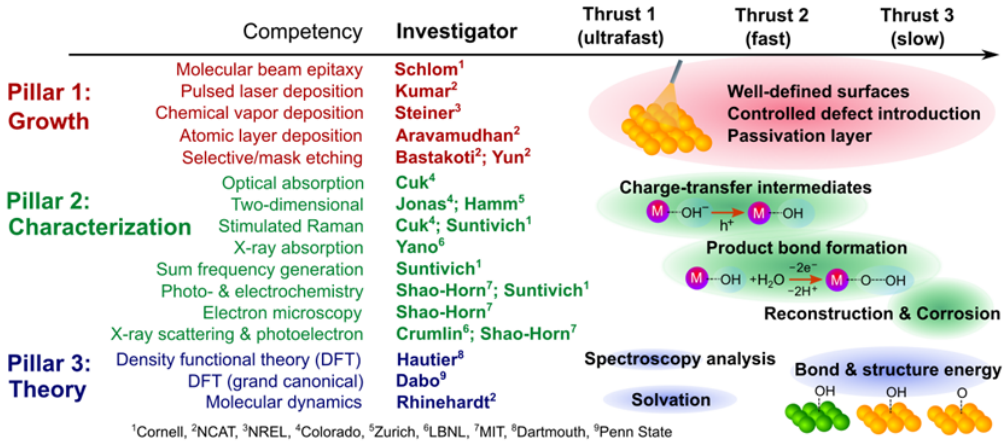


Fig.1: CEDARS team and intertwined three research thrusts.

The research in Thrust 3 is focused on establishing a connection between catalytic turnover, surface restructuring, 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)	
North Carolina Agricultural and Technical State University	Dhananjay Kumar (Director), Kristen Rhinehardt (Deputy Director), Shyam Aravamudhan, Bishnu Bastakoti, Yeoheung Yun
Cornell University	Jin Suntivich, Darrell Schlom
Dartmouth College	Geoffroy Hautier
LBNL	Ethan Crumlin, Junko Yano
MIT	Yang Shao-Horn
NREL	Myles Steiner
Carnegie Mellon University	Ismaila Dabo
University of Colorado at Boulder	Tanja Cuk (Associate Director), David Jonas

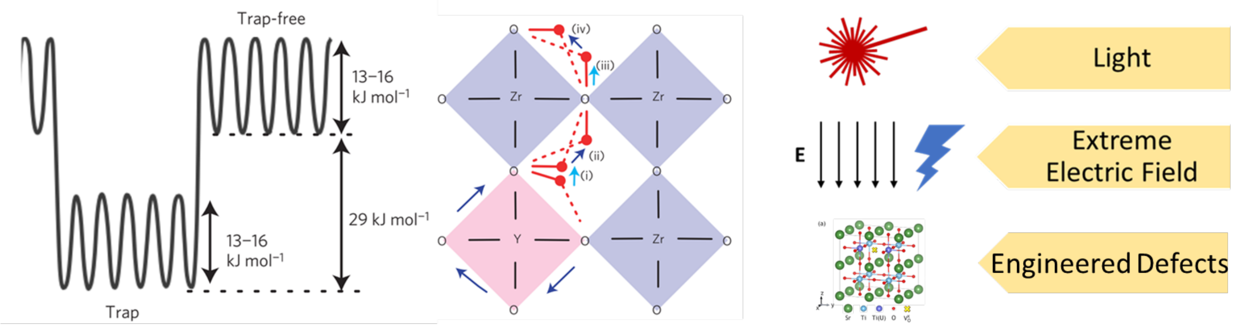
Contact: Dhananjay Kumar, Director, dkumar@ncat.edu
336-2850-3227, https://www.cedars-ncat.org/

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 exceptionally **high electric fields** and **photon** illumination, where the latter additionally induces **phonon** modulation. These external drivers couple to the energy landscape of hydrogenic species and can serve as a means of engineering higher dimensional defects, can alter the dominant charge state of H and distribution within trap sites, and can also drive transport and reaction dynamics at strongly nonlinear field-strength regimes.

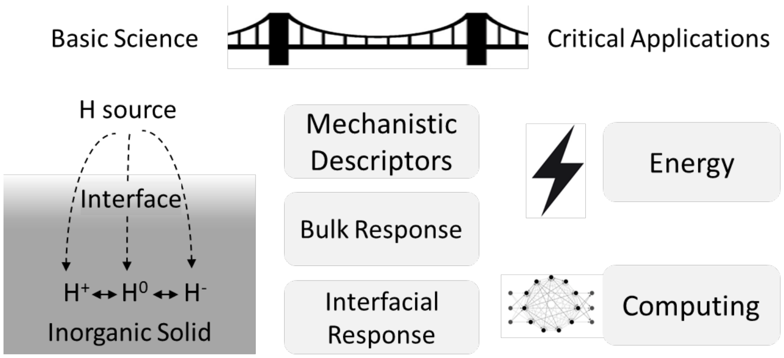


Beyond its special place in physical chemistry, hydrogen is of tremendous societal importance in energy technologies and of growing importance in energy-efficient computing. In both arenas, the relevant devices are limited by hydrogen kinetics, whether it be electrochemical reaction at an interface or 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 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

Contact: William Kung, Senior Director, Operations and Strategic Planning, w-kung@northwestern.edu
847-491-4961, https://heiss.northwestern.edu

Center for Molecular Quantum Transduction (CMQT)

EFRC Director: Michael R. Wasielewski
Lead Institution: Northwestern University
Class: 2020 – 2028

Mission Statement: *To develop the fundamental scientific understanding needed to conduct quantum-to-quantum transduction through a bottom-up synthetic approach that imparts atomistic precision to quantum systems.*

Molecular architectures provide unmatched flexibility for tailoring spin properties, electronic properties, and geometry, which are essential for interfacing disparate quantum degrees of freedom. Molecular synthesis allows bottom-up construction of novel materials with structural reproducibility and modularity, atomic scale spatial control, and access to uniquely molecular degrees of freedom that can be used to conduct spin-photon and spin-spin transduction. Thus, **CMQT** research on quantum-to-quantum transduction responds directly to PRO 3 of the DOE Report of the Basic Energy Sciences Roundtable on *Opportunities for Basic Research for Next-Generation Quantum Systems*. **Key hypothesis:** Quantum information can be transferred coherently between the quantum degrees of freedom available in molecules to access new functionality for quantum information science (QIS).

CMQT is uniquely positioned to exploit recent breakthroughs from our team including landmark coherence times and stabilities of molecular qubits, new quantum materials, as well as the ability to create hybrid qubits and resonant photonic architectures. Moving 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 will also leverage innovative 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. These tools include ultrafast optical and microcavity techniques, pulsed microwave and optically detected magnetic resonance (ODMR), ultrafast low temperature measurements using superconducting resonators, and x-ray structural techniques. Theory approaches include electronic structure, spin dynamics, and exciton dynamics calculations.

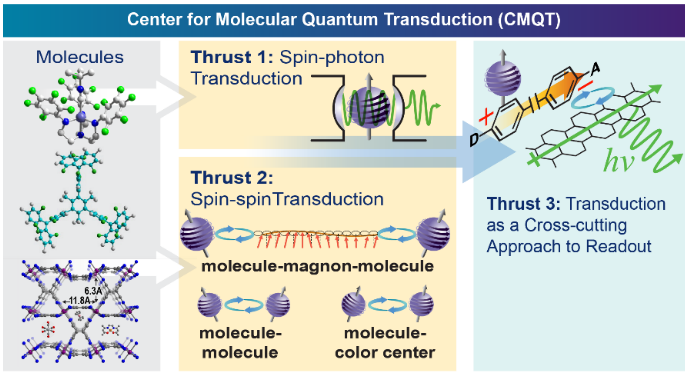


Fig. 1. Overview of **CMQT** research.

Achieving molecular quantum-to-quantum transduction is necessarily an interdisciplinary effort, requiring the scope of an EFRC to assemble the needed expertise for design and synthesis of molecular and solid-state materials, measurement of coherent quantum states at the single quantum level, and seamless incorporation of theory and modeling of materials and measurements. **CMQT**'s team of chemists, physicists, and materials scientists has been assembled with exactly this challenge in mind. Each team member brings a suite of tools to bear that have been extensively validated by our current **CMQT** research, and a history of successful collaborations with one another as evidenced by joint publications.

Thrust 1. Molecular Spin-Photon Quantum Transduction (co-Leaders: Goldsmith and Fuchs). *The goal of Thrust 1 is to develop quantum-to-quantum transduction between molecular spins and photons, spanning* *organic and hybrid materials, with photons spanning visible, telecom, and microwave frequencies.* Strong interactions are necessary for quantum transduction. During 2020-2024, **CMQT** established spin-photon interactions in (1) molecular spin materials coupled to microcavities, and (2) molecular color centers, which are molecular analogues to diamond nitrogen vacancy (NV) centers where spin determines photon emission. During 2024-2028, **CMQT** will build on these platforms and use spin-photon coupling to link spin-spin transduction platforms from Thrust 2 with photons for wider transmission (**Fig. 1**).

Thrust 2. Molecular Spin-Spin Quantum Transduction (co-Leaders: Johnston-Halperin and Long). *The goal of Thrust 2 is to explore the transfer of angular momentum in spin-spin interactions to develop a modular and generalized approach to transducing quantum information between dissimilar quantum systems.* We will explore both local, e.g., qubits within the same molecule or between coupled molecular-solid state qubits, and distributed, e.g., spin-magnon and spin-magnon-spin, coupling to understand and control flow of quantum information. Work will focus on three regimes of spin-spin coupling: (1) transduction between paramagnetic molecular spins and excitations of ferrimagnetically ordered materials like vanadium tetracyanoethylene, $V(TCNE)_x$, (magnons or spin-waves), (2) transduction between paramagnetic electron spins within extended molecular systems, and (3) transduction between paramagnetic spins on different materials, e.g., molecules and solid-state color centers (**Fig. 1**).

Thrust 3. Molecular Transduction as a Cross-cutting Approach for Readout (co-Leaders: Stern and Flatté). *The goal of Thrust 3 is to pioneer new strategies for employing transduction to enhance readout of quantum information contained in molecular spin qubits.* By deploying approaches from Thrusts 1 and 2, **CMQT** will apply two distinct but complementary methods using transduction between molecular spin and charge states to facilitate readout: (1) transducing a spin qubit to a different spin-sensitive host whose state can be more easily read out, and (2) converting a spin qubit to a charge state accessible for electrical readout, which can potentially be compatible with on-chip quantum information platforms. In both approaches, transduction in or between molecules is a key step that enables new measurement schemes (**Fig. 1**) to address the important and pervasive problem of readout of quantum systems.

Center for Molecular Quantum Transduction (CMQT)	
Northwestern University	Michael Wasielewski (Director), Mark Hersam, Matthew Krzyaniak, George Schatz, Nathaniel Stern, Roel Tempelaar, Ryan Young (Director of Operations)
Princeton University	Nathalie de Leon
University of Iowa	Michael Flatté
Massachusetts Institute of Technology	Danna Freedman
Cornell University	Gregory Fuchs
University of Wisconsin-Madison	Randall Goldsmith
Ohio State University	Ezekiel Johnston-Halperin
University of California, Berkeley	Jeffrey Long
Rice University	Xuedan Ma
University of Chicago	Peter Maurer
University of California-San Diego	Joel Yuen-Zhou

Contact: Michael R. Wasielewski, Director, m-wasielewski@northwestern.edu
847-467-1423, http://cmqt.org

Fast and Cooperative Ion Transport in Polymer Based Materials (FaCT)
EFRC Director: Valentino 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.*

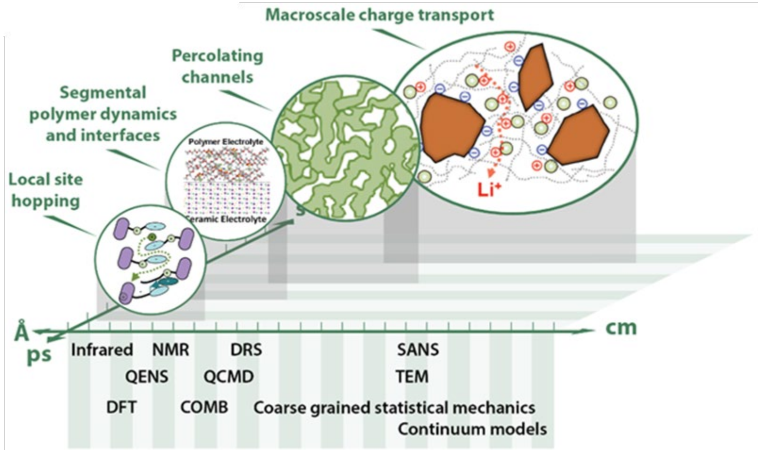


Figure 1 Length and time scales of experiments and theory relevant to the FaCT mission. (QENS = quasielasitc neutron scattering, BDS = broadband dielectric NMR = nuclear magnetic resonance, DFT = density functional theory, DRS = diffuse reflectance spectroscopy, QCMD = quartz crystal microbalance with 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?

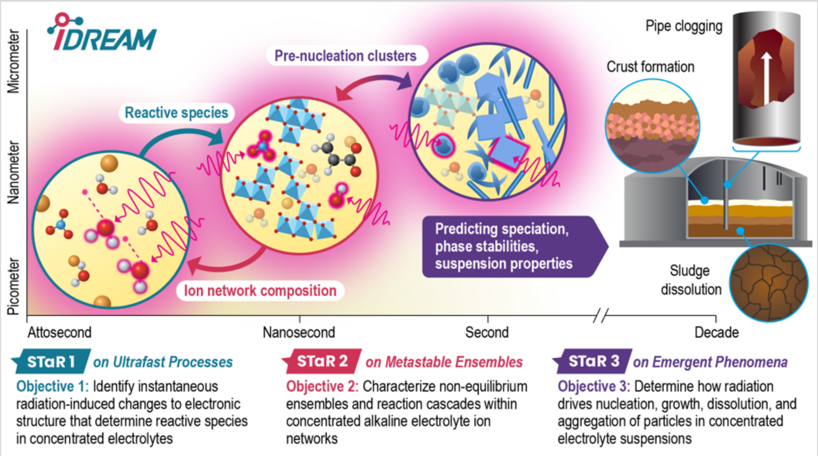
Crosscut: Design through computation, theory, and materials informatics. The crosscut goal is to leverage fundamental knowledge obtained from the Thrusts to enable the design of fast ion conducting polymer and polymer-ceramic electrolytes. ***Key questions***: How can representation learning be utilized 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 Cooper (Director), Chelsea (Xi) Chen (Thrust 2; Co-lead), Sheng Dai, Catalin Gainaru, Rajeev Kumar (Thrust 1; Co-lead), Tomonori Saito, Yuya Shinohara, Alexei Sokolov (Thrust 2; Co-lead), Thomas (Zac) Ward, Albina Borisevich
The Pennsylvania State University	Susan Sinnott (Deputy Director), Ralph Colby, Wesley Reinhart (Crosscut; Co-lead)
University of California, Santa Barbara	Raphaële Clément
University of Tennessee, Knoxville	Takeshi Egami
Michigan State University	Michael Hickner (Thrust 1; Co-Lead)
Texas A&M University	Jodie Lutkenhaus (Thrust 1; Co-Lead)
University of Illinois Urbana-Champaign	Kenneth Schweizer
Georgia State University	Gangli Wang

Contact: Sadie Spicer, Managing Director, sco3@psu.edu
(814) 863-0525, https://fact-efrc.ornl.gov

Ion Dynamics in Radioactive Environments and Materials (IDREAM)
EFRC Director: Carolyn Pearce
Lead Institution: Pacific Northwest National Laboratory
Class: 2016 – 2028

***Mission Statement**: To master the cascade of radiation chemistry that drives far from equilibrium speciation and reactivity in chemically complex environments, linking attosecond timescales to decadal processes.*



Objective 1: Identify instantaneous radiation-induced changes to electronic structure that determine reactive species in concentrated electrolytes

Objective 2: Characterize non-equilibrium ensembles and reaction cascades within concentrated alkaline electrolyte ion networks

Objective 3: Determine how radiation drives nucleation, growth, dissolution, and aggregation of particles in concentrated electrolyte suspensions

Plutonium production at the Department of Energy's Hanford Site has resulted in the most challenging environmental cleanup in history. Retrieval of 200 million liters of waste containing 170 million curies of radioactivity from underground storage tanks will take billions of dollars and decades of time. Waste cleanup challenges stem primarily from supernatant, salts, and sludges containing high concentrations of sodium hydroxide, aluminate, nitrate, and nitrite, with co-disposed organics, phosphate, and trace metals. The extreme alkalinity, low water content, and continuous production of transient reactive species by ionizing radiation from radionuclides in the waste drives chemistry far from equilibrium, as indicated by a plethora of oxidized and reduced species that exist simultaneously. The extreme conditions also result in poorly predictable dissolution and reprecipitation behavior of the aluminum (oxy)hydroxides that form the bulk of the waste solids.

This is a critical moment for the tank waste cleanup mission, with the start-up of the Waste Treatment and Immobilization Plant to vitrify low-activity liquid waste. Sludge processing is not expected to start for 10 years. Given the long timeframe, a fundamental understanding of how the radiation environment changes waste speciation and reactivity is essential for developing effective and efficient processing strategies. With waste retrieval contractors, IDREAM identified where continuous radiation-induced creation of reactive species has an important influence on tank waste evolution: (1) radiolysis of water, organics, and nitrogen species, resulting in changes in pH, and creation of flammable and/or toxic gases; (2) initiation of redox reactions that convert nitrate to nitrite and affect technetium, iron, and chromium speciation; and (3) changes to mineral dissolution and precipitation rates. The advent of powerful attosecond and femtosecond X-ray pulses synchronized with a variety of ultrafast probes ushers in a new era for radiation chemistry to help unravel the complexities of Hanford Site tank waste.

Previous accomplishments uniquely position IDREAM to now focus on how radiation has driven solution speciation and precipitation to yield the complex phase assemblages at the heart of the waste processing challenge. IDREAM research already has made specific, tangible impacts on the Hanford Site cleanup mission by advancing fundamental knowledge in understanding and controlling chemical and physical processes at interfaces. For example, it is now known that gibbsite supersaturation in tank waste is related to the presence of ion networks in solution.

By addressing our project objectives with combined experimental and computational expertise, IDREAM research will transcend many orders of magnitude of spatiotemporal resolution to tackle the problems related to far from equilibrium conditions that result from the interplay between high ionic strength and radiation-induced metastability in radioactive tank waste.

These four-year project objectives will elucidate the radiation-driven evolution of reactive species and precipitates that emerge from complex ion networks across three SpatioTemporal Regimes (STaRs) to:

1. Identify the instantaneous, radiation-induced changes to electronic structure that determine reactive species in concentrated electrolytes. STaR1 on Ultrafast Processes (attoseconds, picometers) will reveal the key reactive species that form upon irradiation of ion networks and follow their local evolution with ultrafast spectroscopies including sub-femtosecond X-ray snapshots.
2. Characterize nonequilibrium ensembles and reaction cascades within concentrated alkaline electrolyte ion networks. STaR2 on Metastable Ensembles (nanoseconds, nanometers) will elucidate the ion-network response to radiation-induced reactive species identified in STaR1 and the role of solution heterogeneity in the formation of metastable ensembles of species that ultimately transition to suspensions and precipitates examined in STaR3.
3. Determine how radiation drives nucleation, growth, dissolution, and aggregation of particles in concentrated electrolyte suspensions. STaR3 on Emergent Phenomena (seconds, micrometers) focuses on radiation-driven phase transformation processes and outcomes that dominate mesoscale observables relevant to tank waste over long time periods, including the composition, physical characteristics, and stabilities of particle suspensions and solids.

The broader scientific impact of IDREAM research will reveal mechanisms by which properties of matter emerge from complex chemical systems in far from equilibrium radioactive environments. For the first time, we will show how limited water and key solutes influence the generation of short-lived transient reactive species in concentrated electrolytes, then how these reactive species govern ion-pair formation, oligomerization, long-range ordering, and the pre-nucleation species that lead to the precipitation of new phases. Our work to understand radiation-induced mechanisms of crystallization will result in innovations in nucleation and crystal growth. Our discovery of impurity effects on critical cluster formation will inform environmental remediation. Beyond radioactive tank waste, our distinctive capabilities, including ultrafast spectroscopy to study radiolysis and multiscale-simulation tools, will have a broad and foundational impact on chemical and materials sciences.

Ion Dynamics in Radioactive Environments and Materials (IDREAM)	
Pacific Northwest National Laboratory	Carolyn Pearce (Director); Jaehun Chun; James De Yoreo; Greg Kimmel (STaR2 Lead); Kevin Rosso (STaR3 Lead); Greg Schenter (STaR3 Lead); Michael Spradling (Operations); Zheming Wang; Xin Zhang
Argonne National Laboratory	Linda Young (STaR1 Lead)
Georgia Institute of Technology	Thomas Orlando
Hunter College, City University of New York	Lynn Francesconi
Oak Ridge National Laboratory	Lawrence Anovitz
University of Notre Dame	Jay LaVerne (Deputy Director)
University of Utah	Aurora Clark (STaR2 Lead)
University of Washington	Xiaosong Li (STaR1 Lead)

Contact: Carolyn Pearce, Director, carolyn.pearce@pnnl.gov
509-372-4875, www.pnnl.gov/idream

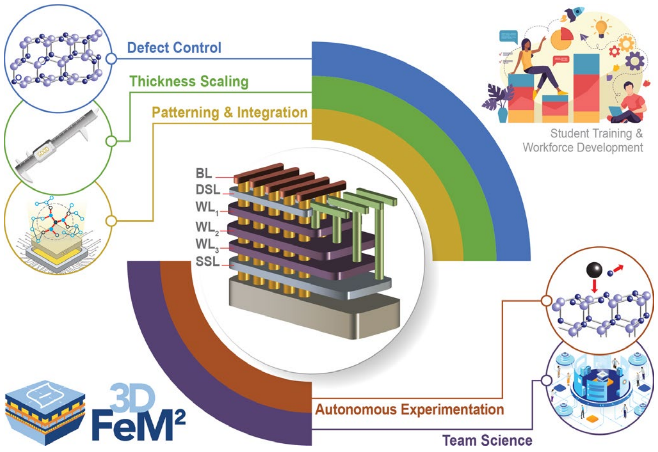
Center for 3 Dimensional Ferroelectric Microelectronics Manufacturing (3DFeM^2)

EFRC Director: Susan Trolier-McKinstry
Lead Institution: The Pennsylvania State University
Class: 2020 – 2028

Mission Statement*: To integrate ferroelectric films at the required scale and reliability for 3D memory using next generation manufacturing practices.*

3DFeM^2 will develop three-dimensional (3D) non-volatile compute-in-memory technology that integrates novel ferroelectric materials into classical semiconductor manufacturing processes. This technology addresses tremendous societal needs for microelectronics that consume less energy without sacrificing performance, circumvents the end of Moore's law in 2D scaling, and overcomes the "von Neumann bottleneck."

3DFeM^2 will enable compute-in-memory chips and smart manufacturing by exploring the fundamental materials-physics of novel ferroelectricity and by developing a deeper scientific understanding of the underlying processes of 3D integration of ferroelectrics. Ultimately, the outcomes will enable a million-fold enhancement in interconnection between memory and logic, along with order of magnitude reductions in the energy cost to computation. ***This will be realized by developing the fundamental manufacturing science that assists the semiconductor transition to the fourth industrial revolution (Industry 4.0), a needed step for integrating robust ferroelectric materials that show outstanding properties throughout the device's lifetime.*** The proposed 3D integrated ferroelectrics–deposited on geometrically complex surfaces with realistic back-end-of-the-line (BEOL) processing–will naturally fuse memory and computation and exploit the 3rd dimension, unleashing unprecedented capabilities for the next generation of computation and artificial intelligence. 3DFeM^2 will employ a data-driven co-design approach that overcomes the enormous obstacles for integrating new materials and realizing the lab-to-fab transition. We will implement digitalized processing instruments to generate large data sets, use physics-based machine learning (ML) to reduce experimental dimensionality, identify descriptors that can be monitored to assess properties in real time, establish autonomous experiments to expedite optimization, and develop digital twins that accelerate innovation through fast, cost-effective virtual experiments. In the process, 3DFeM^2 will transform semiconductor manufacturing practices while solving a critical societal energy challenge.



Vision for 3DFeM^2 in which fundamental materials science and processing science enable 3D ferroelectric memory for low power computing.

Ferroelectric fluorite and wurtzite crystals fundamentally change the device and integration landscape for ferroelectric microelectronics. *One of the key challenges for integrating new ferroelectrics is identification of the links between processing pathways and defects across length scales*. The role of defects must be understood early in the process of assessing new materials so potential failure modes can be identified prior to expensive scale-up in foundry settings. Key strategies for 3DFeM^2 are: ***i.*** innovations in materials and 3D structures that enable high-performance 3D ferroelectric random-access memory and high-density 3D ferroelectric NAND storage solutions; ***ii.*** materials-device-manufacturing co-design and co-optimization informed by ML models based on *in situ* processing and characterization data; and ***iii.*** digital twin creation of 3D ferroelectric memory to significantly accelerate the R&D to manufacturing pipeline.

The 3DFeM^2 will target the following science goals: ***i.*** Understand how materials-chemistry design rules and processing boundary conditions regulate ferroelectricity in novel materials, ***ii.*** Understand and control ferroelectricity-enabling as well as the lifetime-limiting defects in next-generation ferroelectrics, ***iii.*** identify synthesis "descriptors," e.g., semi-empirical observables for defect chemistry, structure, and property evolution during material growth and etching, ***iv.*** characterize materials at previously inaccessible time and length scales, utilizing ML and *in situ* microscopy to identify phases, switching mechanisms, and scaling trends, ***v.*** enable autonomous workflows for optimized film growth and etching in 3D, ***vi.*** Generate device concepts best aligned with novel ferroelectric properties and target property metrics that maximize performance, and ***vii.*** establish a digital twin workflow that predicts performance and reliability with high accuracy based on device configuration, manufacturing process history and system-level operational characteristics.

3DFeM^2 will test two overarching hypotheses:

(1) Understanding domain walls, defects, interfaces, and intermediate switching structures of next-generation ferroelectrics at multiple length-scales will enable 3DFeM^2 to engineer their coercive field, leakage current, and endurance characteristics and to discover superior ferroelectric formulations.

(2) Combining ML, *in situ* monitoring during synthesis and etch, and *in operando* device measurements will inform manufacturing science and expedite co-development of novel materials into 3D systems with high yields while creating a blueprint for future process innovations

Center for 3 Dimensional Ferroelectric Microelectronics Manufacturing (3DFeM^2)	
The Pennsylvania State University (lead)	Susan Trolier-McKinstry (Director) Betul Akkopru-Akgun, Venkatraman Gopalan, Thomas N. Jackson, Ying Liu, Jon-Paul Maria, Vijaykrishnan Narayanan, Darren Pagan, Clive Randall, Adri van Duin
Carnegie Mellon University	Elizabeth Dickey
Georgia Institute of Technology	Suman Datta, Lauren Garten, Asif Khan
The University of Notre Dame	Kai Ni
University of Maryland	A. Gilad Kusne, Ichiro Takeuchi
University of Pennsylvania	Andrew Rappe
University of Tennessee - Knoxville	Sergei Kalinin
University of Virginia	Patrick Hopkins, Jon Ihlefeld
Sandia National Laboratories	M. David Henry
Brookhaven National Laboratory	Judith Yang
Oak Ridge National Laboratory	Kyle Kelley, Rama Vasudevan

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

Bio-inspired Light-Escalated Chemistry (BioLEC)

EFRC Director: Gregory Scholes
Lead Institution: Princeton University
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.*

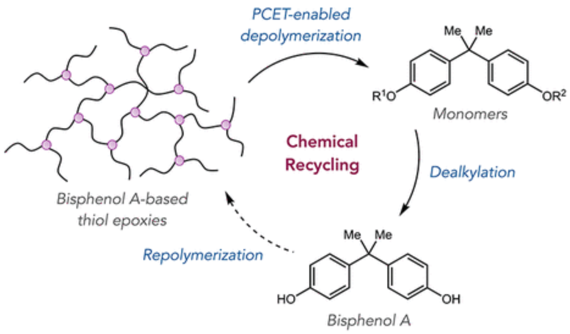
The search for more efficient, cost-effective, and green manufacturing technologies is of paramount importance. Chemical manufacturing accounts for 10% of total global energy consumption and is responsible for 7% of the world's greenhouse gas emissions. Fossil fuels remain the most widely employed source 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 to perform 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. In this second phase of our center, we advance towards our goals through three thrusts:

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

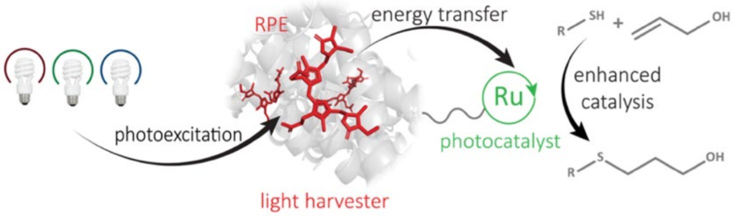
We develop new photochemistry and photochemical feedstocks in BioLEC, and expand the scope of mechanistic tactics and reactive substrates available to the photocatalysis community. Drawing on the rich physical chemistry capabilities in BioLEC, new photochemical techniques for reactivity and spectroscopic characterization are developed. New reactivities push the boundaries of photoexcitation and accessible redox potentials. Finally, we find ways to use this new chemistry to valorize feedstocks that were previously inaccessible using photochemistry, providing a pathway toward recycling waste.



Chemical recycling of thiol epoxy thermosets enabled by light-driven O-H proton-coupled electron transfer

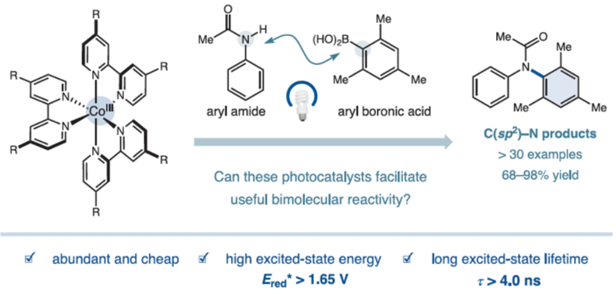
B. Emulate or exploit biological systems for new or improved syntheses

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 introduce functionality that first mimics and then exceeds that of natural systems. Additionally, bioinspired tactics from photosynthesis such as light harvesting take advantage of separating energy capture and catalytic functions to improve efficiency and/or selectivity.



Light-harvesting protein conjugated to Ru photocatalyst increases yields and enables reactions at previously unusable irradiation wavelengths

C: Inform design of photocatalysts by elucidating photocatalysis mechanisms



Marcus inverted long-lived highly oxidizing cobalt photocatalysts

We advance fundamental knowledge of the elementary organometallic steps available through photoexcitation by applying a variety of physical techniques (e.g., transient absorption spectroscopy, organometallic synthesis, EPR, pulse radiolysis, ultra-fast IR, as well as newly developed techniques) to study the mechanisms in multiple metallaphotoredox reactions. We have built on these efforts with the goal of gaining a more global understanding of how changes to substrate, ligand, photocatalyst, and other reaction factors affect the reaction mechanism. This detailed mechanistic understanding enables rational catalyst design, for instance in adopting more earth-abundant metal catalysts instead of expensive rare earth complexes.

Bioinspired Light-Escalated Chemistry (BioLEC)	
Princeton University	Gregory Scholes (Director), Paul Chirik, Todd Hyster, 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
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
University of California, Los Angeles	Abigail Doyle
University of Colorado Boulder	Obadiah Reid

Contact: Victoria Cleave, Managing Director, vcleave@princeton.edu
609-258-7020, https://biolec.princeton.edu

Quantum Photonic Integrated Design Center (QuPIDC)
EFRC Director: Libai Huang
Lead Institution: Purdue University
Class: 2024 – 2028

***Mission Statement**: to discover, design, and realize robust many-body entangled photon and matter states through multi-scale co-designing strategies in heterogeneous solid-state photonic systems.*

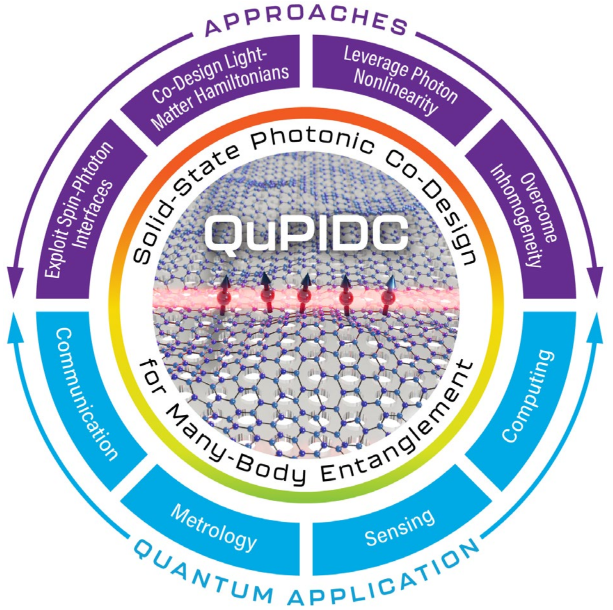


Figure 1. **QuPIDC** targets the realization of resilient many-body entangled states of photons and matter. The illustration depicts the entanglement of spins from multiple emitters in a h-BN layer with a disordered energy landscape and coupled to a photonic crystal.

Photonic platforms based on solid-state materials present a promising path toward the scalable generation of complex quantum states of light, which are essential for the development of quantum networks, entanglement-enhanced metrology, and optical quantum computation. The key enabler of these technologies is the efficient and reliable generation of quantum light. Solid-state quantum emitters—including defects, quantum dots, molecules, and two-dimensional (2D) materials—have demonstrated significant potential for generating quantum light, while advances in nanophotonic structures have made it possible to manipulate light at the chip level.

However, despite the progress, significant scientific challenges remain, particularly regarding scalability. Inhomogeneities and dephasing, which are inherent to solid-state systems, have limited the generation of quantum light to small photon numbers. Addressing noise channels, such as decoherence and inhomogeneity, is essential for advancing quantum photonic systems. Moreover, new strategies are required to generate and characterize robust, many-photon entangled states in the presence of imperfections.

The **Quantum Photonic Integrated Design Center (QuPIDC)** aims to systematically address these challenges by leveraging the complexity and imperfections of solid-state materials to discover, design, and realize robust many-body entangled photon and matter states. This will be achieved through a multi-scale co-design strategy that integrates solid-state quantum emitters with advanced nanophotonic structures. The QuPIDC team is uniquely equipped for this mission, with expertise spanning solid-state quantum emitters, nanophotonics, materials growth, quantum optics, ultrafast spectroscopy, and quantum many-body dynamics.

QuPIDC's Goals
Over the next four years, QuPIDC will pursue three primary goals:

1. **Co-design Quantum Emitters and Nanophotonic Structures**: The center will integrate quantum emitters with nanophotonic structures to reduce decoherence, increase photon collection

efficiency, and enable control over spin and polarization. This integration will also enhance nonlinear interactions crucial for generating quantum light.

2. **Prepare Entangled Photon and Emitter States in Inhomogeneous Systems**: Using Hamiltonian engineering and quantum control strategies, QuPIDC will generate entangled states of photons and emitters within complex, inhomogeneous systems. This approach will allow the creation of robust quantum states despite intrinsic material imperfections.
3. **Develop New Theoretical and Experimental Tools**: QuPIDC will create innovative theoretical frameworks and experimental techniques to identify, generate, and characterize complex quantum states of light and matter. These tools will be essential for realizing many-photon quantum states that are resilient to noise and loss.

Key Research Areas

To achieve these goals, QuPIDC will focus on several key areas of research, which include:

- **Nanophotonic Structure Design**: The center will design dielectric and plasmonic nanophotonic structures that enhance the optical and spin coherence of quantum emitters, including h-BN, transition metal dichalcogenides (TMDs), Cu_2O, GaAs quantum dots, organic molecules, and color centers in SiC, SiN, and diamond.
- **Co-designing Light-Matter Interactions**: QuPIDC will co-design photon-emitter interfaces to maximize nonlinear photon interactions, critical for increasing photon entanglement. These enhanced light-matter interactions will enable the realization of highly entangled photon states, including cluster states, superradiance, Fock states, and squeezed light.
- **Quantum Control and Characterization**: Theoretical and experimental techniques will be developed to control and characterize entangled photon states and multi-emitter systems.

Alignment with BES Research Priorities

QuPIDC's research strategies align with the **Priority Research Directions (PRDs)** and **Priority Research Opportunities (PROs)** outlined in the Basic Energy Sciences (BES) workshop and roundtable reports. Specifically, QuPIDC's emphasis on the co-design of quantum emitters and nanophotonic structures directly supports the transformative materials paradigm for quantum information science. The center's development of a comprehensive suite of theoretical and simulation tools will address bottlenecks in experimental preparation and characterization of complex quantum states.

Quantum Photonic Integrated Design Center (QuPIDC)	
Purdue University	Libai Huang (Director), Alexandra Boltasseva (Deputy director), Hadiseh Alaeian, Jonathan Hood, Tongcang Li, Vlad Shalaev, Valentin Walther
Los Alamos National Laboratory	Han Htoon, Sergei Tretiak
Northwestern University	Teri Odom
Stanford University	Jelena Vučković
University of Chicago	Jiwoong Park
University of Maryland Baltimore County	Matthew Pelton
University of Oklahoma	Alisa Javadi
Virginia Tech University	Sophia Economou
North Carolina State University	Sabre Kais

Contact: Libai Huang, Director, libai-huang@purdue.edu
765-494-7851, https://www.science.purdue.edu/qupidc/

Center For Energy Efficient Magnonics (CEEMag)
EFRC Director: Yuri Suzuki
Lead Institution: SLAC
Class: 2024 – 2028

***Mission Statement**: To advance the basic scientific understanding of magnon excitation, propagation, transduction, and control that is motivated by an end use of magnon-based interconnects and their integration into microelectronics.*

Magnons provide the promise for microelectronics with low-loss information and energy transfer at the nanoscale using propagating excitations with wavelengths that are orders of magnitude smaller than microwave or photonic interconnects. Fundamental innovations in materials, theory, and structures for magnonic systems in a co-design approach will enable new THz-frequency, low-loss interconnects based on spin wave excitations that can also deliver active control including switching, modulation, and amplification. Not only can information be encoded in both amplitude and phase of magnons, but also nonlinearity and non-reciprocity can provide additional functionality, making magnons far more versatile than electrons in future interconnects. We will exploit THz frequencies characteristic of strong exchange interactions in ferrimagnets (FiM) and antiferromagnets (AFM), focusing on low-damping semiconducting and insulating FiMs and AFMs to minimize charge current dissipation. Low-loss oxides will be integrated into silicon-based platforms via co-design of interconnect characterization, nanofabrication, and theory. CEEMag will build on previous scientific and commercial success in spin-based microelectronics, including spin torque phenomena for memory applications pioneered by several team members.

In order to develop a comprehensive understanding of magnon excitation, propagation, transduction and control for realizing magnon interconnects (*Figure 1*), we will:

- manufacture resilient FiM and AFM materials with record low damping
- demonstrate *robust* tunability of FiM and AFM materials to functionalize magnon interconnects
- enhance magnon transmission within and among FiM and AFM materials as well as at interfaces which can dominate insertion losses
- generate and detect coherent and incoherent magnons efficiently from GHz to THz frequencies and identify tradeoffs in interconnect usage
- develop a framework for nonlinear magnon behavior permitting *robust* magnon control

To meet these four-year integrated scientific goals of CEEMag, we have assembled an EFRC team with experience in materials synthesis, local and element-specific characterization of spin phenomena, THz and GHz spectroscopy, device design and nanofabrication, along with theoretical expertise in materials design, spin transport, and device architecture. The materials growth experts, who bring a range of techniques for complex oxides and 2D materials, will work in tandem with theorists in materials design and with characterization experts to achieve low damping and enhanced magnon transmission in *manufacturing-*

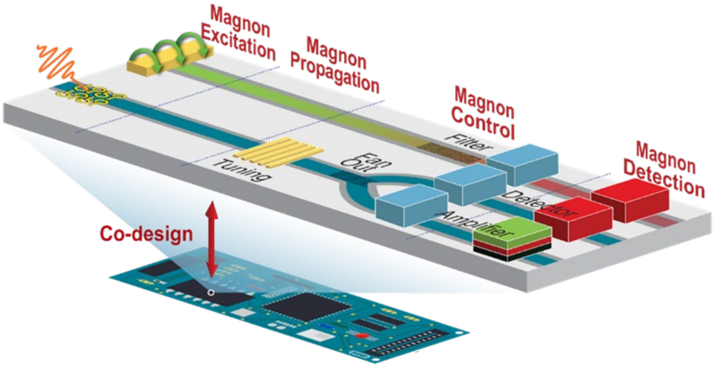


Figure 1. EFRC Conceptualization. Magnon interconnects will be realized by addressing fundamental questions concerning magnon excitation, propagation, control and detection within three research thrusts.

resilient FiM and AFM materials. We will realize interconnect structures to enhance magnon transmission within FiM and AFM materials as well as at interfaces which can dominate insertion losses. Team members will demonstrate energy-efficient magnon detection from GHz to THz frequencies and mechanisms for *robust* tunability to enable switches, modulators, and amplifiers. Theoretical simulations will be integrated with experiments in all of these efforts. Theoretical efforts will also develop a framework for beneficial utilization of magnon nonlinearities and to identify tradeoffs affecting performance.

To tackle the multi-disciplinary scientific challenges of understanding magnon behavior and making useful interconnects, we have organized CEEMag into three interconnected research thrusts:

> **THRUST 1: ADVANCED MATERIALS FOR MAGNON INTERCONNECTS** identifies promising magnon materials and how best to excite and visualize magnons
>
> **THRUST 2: ENERGY EFFICIENT MAGNON DETECTION** addresses the challenges associated with transducing GHz-THz magnons to electrical signals
>
> **THRUST 3: FUNCTIONALITY IN MAGNON INTERCONNECTS** addresses the fundamental limits associated with control of magnon amplitude, propagation path, and interference between their excitation and detection

Our scientific discoveries related to magnon interconnects will enable critical elements such as sources, waveguides, amplifiers, mixers, and devices based on non-reciprocity. By examining new materials, magnon phenomena (e.g., excitation, detection, frequency conversion, nonlinearities) and their practical implementation, we will not only address the challenges associated with magnon excitation, transmission, detection and control but also develop magnonic interconnects with functionality beyond simple transduction. Throughout the EFRC, the projects will adopt a co-design approach of theory, materials, and characterization.

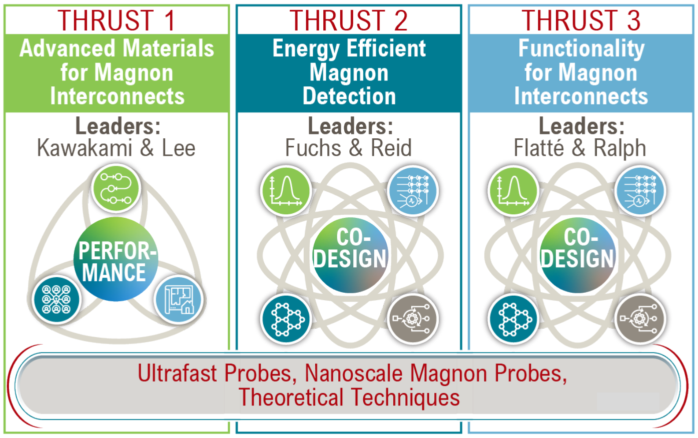


Figure 2. CEEMag organization. Each of the three research thrusts are comprised of a co-design approach to achieve the goal of realizing magnon interconnects along with cross-cutting themes of ultrafast probes, nanoscale magnon probes and theoretical techniques.

Center For Energy Efficient Magnonics (CEEMag)	
SLAC	Yuri Suzuki (Director), Alexander Reid (Deputy Director), Georgi Dakovski, Matthias Hoffmann Harold Hwang, Wei-Sheng Lee
Cornell University	Gregory Fuchs, Daniel Ralph
Morgan State University	Ramesh Budhani
Northwestern University	Pedram Khalili
Ohio State University	Roland Kawakami
University of California, Irvine	Clare Yu
University of Iowa	Michael Flatté, Durga Paudyal
University of Texas, Austin	Elaine (Xiaoqin) Li

Contact: Yuri Suzuki, Director, ysuzuki1@stanford.edu
(650) 724-4007, https://ceemag.slac.stanford.edu

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 EFRC 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**).

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.

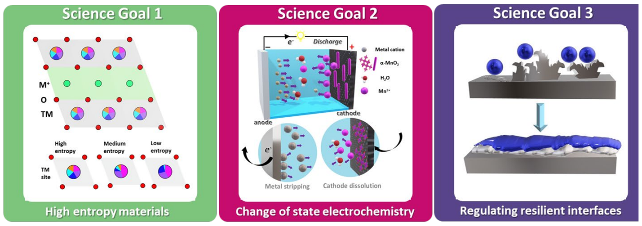


Figure 1. *m2m#S* Center Science Goals (SG).

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: $\Delta G_{mix} = \Delta H_{mix} - T\Delta S_{mix}$, where ΔG_{mix}, ΔH_{mix}, and ΔS_{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)	
Stony Brook University	Amy Marschilok (Director); Kenneth Takeuchi (Deputy Director); Esther Takeuchi (Founding Director); Yu-chen Karen Chen-Wiegart; Carlos Colosqui; Stanislaus Wong
Brookhaven National Laboratory	Shan Yan (Center Operations Officer); Ping Liu; Yimei Zhu
Columbia University	Alan West
Cornell University	Lynden Archer
Lehigh University	Elsa Reichmanis
University of Texas, Austin	Guihua Yu
Xavier University of Louisiana	Lamartine Meda

Contact: Amy Marschilok, Director, amy.marschilok@stonybrook.edu
(631)632-8364, https://www.stonybrook.edu/commcms/m2m/

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.*

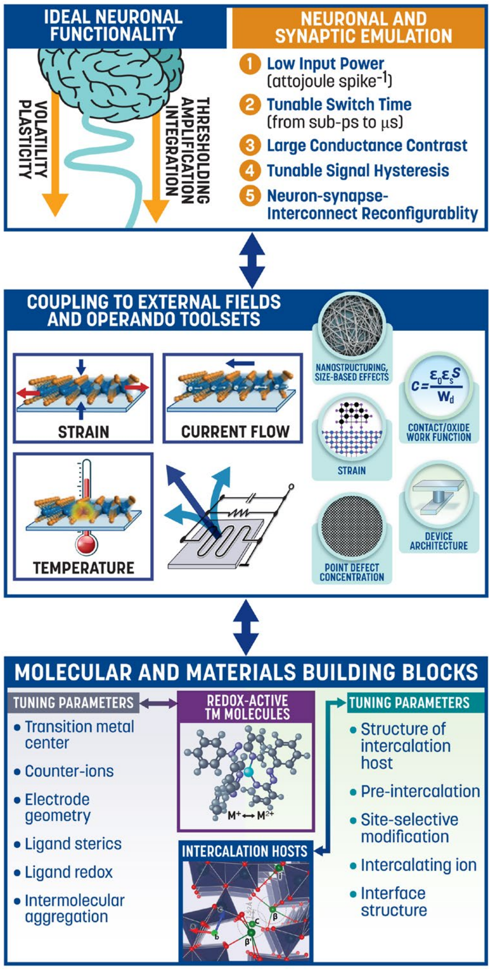


***Fig. 1. REMIND** will create materials for artificial neurons and synapses by controlling the physics and chemistry of nonlinear dynamical electronic transitions to provide a fundamental basis for neuromorphic computing.*

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,

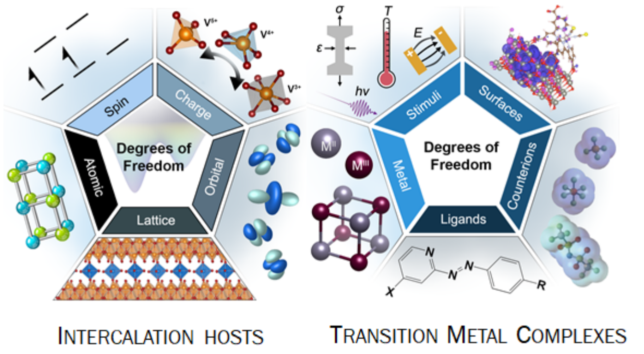


Fig. 2. REMIND *will explore, expand, and navigate the design space of electronic instabilities using materials design, host-guest chemistry, site-selective modification, and lattice strain.*

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.

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
Lawrence Berkeley National Laboratory	Jinghua Guo, Eli Rotenberg
Sandia National Laboratory	Suhas Kumar, Alec Talin

Contact: Sarbajit Banerjee, Executive Associate Director, banerjee@chem.tamu.edu
979-862-3102, https://www.remind.engr.tamu.edu/

Center for Soft PhotoElectroChemical Systems (SPECS)
EFRC Director: Neal Armstrong
Lead Institution: University of Arizona
Class: 2022 – 2026

Mission Statement*: To understand the factors controlling charge and matter transport processes in inexpensive, scalable, and durable π-conjugated polymer (plastic) materials by exploring 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 is fulfilling 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.

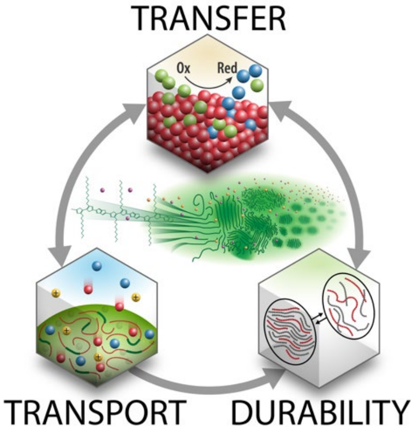


Figure 1. SPECS thrusts working synergistically to understand charge and matter transport processes in soft materials.

Direct outcomes of SPECS are 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 are advancing 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, Hub) programs and Earthshot Initiatives. 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 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 is designing and increasing understanding of π-conjugated polymer (photo)electrodes 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 from photocathodes. Data from theory/modelling, spectroscopy, and structural characterization has been and will continue to 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 tailors 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 is establishing 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.

Center for Soft PhotoElectroChemical Systems (SPECS)	
University of Arizona	Neal Armstrong (Director), Jean-Luc Bredas, Jen Garcia (Program Coordinator)
National Renewable Energy Laboratory	Elisa Miller (Associate Director of Operations), Garry Rumbles (Senior On-Site Advisor), Annie Greenaway, Andrew Ferguson
University of Colorado, Boulder	Seth Marder (Durability Thrust Lead), Mike Toney, Steve Barlow, Obadiah Reid
Emory University	Tianquan (Tim) Lian (Charge Transfer Thrust Lead)
University of Kentucky	Chad Risko (Charge Transport Thrust Lead)
Georgia Institute of Technology	Erin Ratcliff (Associate Director of Scientific Continuity), Natalie Stingelin
Stanford University	Alberto Salleo

Contact: Erin Ratcliff, SPECS Associate Director, eratcliff8@gatech.edu
http://specs.arizona.edu/

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.*

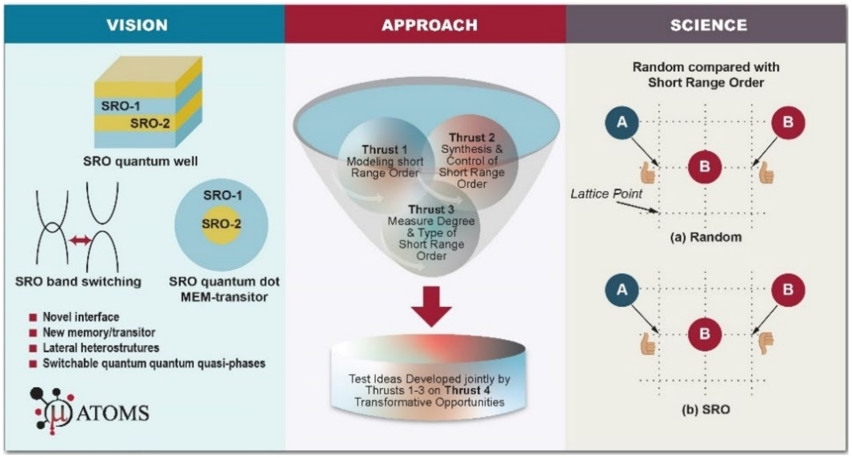


Figure 1 Vision: Precision manipulation of the spatial correlation among atoms as opposed to composition or dopants, to fabricate semiconductor properties using only one composition. **Approach:** Thrust 1-3 integrated into crosscutting Thrust 4 to demonstrate ability to design semiconductor properties 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.

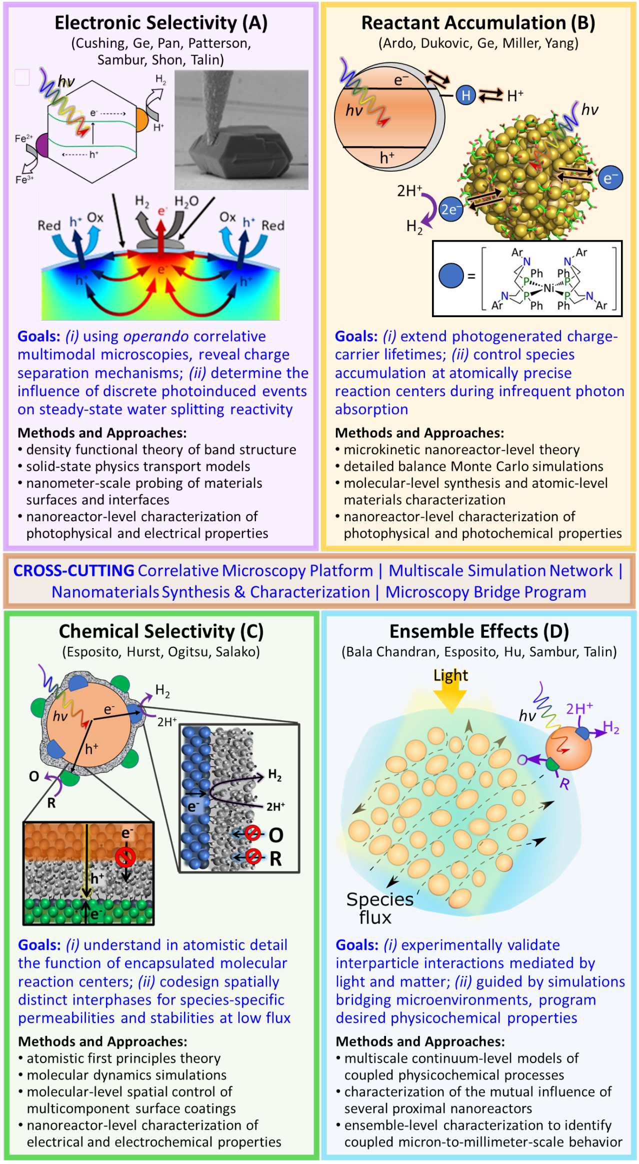
µ-ATOMS	
University of Arkansas	Shui-Qing Yu, Gregory Salamo, Jin Hu, Hugh Churchill, Hiroyuki Nakamura
Sandia National Laboratory	Ezra Bussmann, Shashank Misra, Tzu-Ming Lu, Michael Lilly
George Washington University	Tianshu Li
Rensselaer Polytechnic Institute	Shengbai Zhang
Dartmouth College	Jifeng 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

Contact: Shui-Qing "Fisher" Yu, Director, syu@uark.edu
479-575-7265, https://efrc.uark.edu

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 (STH) efficiency of ensembles of photosynthetic nanoreactors be increased by more than an order-of-magnitude to outperform state-of-the-art photoelectrochemical (PEC) devices*? The key discovery that supports a pathway to answering this question comes from **EPN** numerical simulations, which indicate that STH efficiencies for ensembles of photosynthetic nanoreactors can exceed those of standard PEC devices. The cause of this efficiency enhancement is low absorbed photon flux per nanoreactor, and thus small fuel-forming reaction rates that result in less overpotential than observed for typical PEC photoabsorbers, combined with the multiplicative output from having many photoabsorbers in an ensemble. Toward achieving its scientific mission, **EPN** is guided by scientific research goals that span 4 thrusts, each with directed research approaches and associated methods (*Fig. 1*). Fundamental knowledge gained is being used to identify the physicochemical properties that in aggregate are responsible for ensemble behaviors, which may lead to transformative pathways to meet the DOE H_2 Shot cost target of $1 per kg-$H_2$.



Electronic Selectivity (A)
(Cushing, Ge, Pan, Patterson, Sambur, Shon, Talin)

Goals: *(i)* using *operando* correlative multimodal microscopies, reveal charge separation mechanisms; *(ii)* determine the influence of discrete photoinduced events on steady-state water splitting reactivity

Methods and Approaches:
- density functional theory of band structure
- solid-state physics transport models
- nanometer-scale probing of materials surfaces and interfaces
- nanoreactor-level characterization of photophysical and electrical properties

Reactant Accumulation (B)
(Ardo, Dukovic, Ge, Miller, Yang)

Goals: *(i)* extend photogenerated charge-carrier lifetimes; *(ii)* control species accumulation at atomically precise reaction centers during infrequent photon absorption

Methods and Approaches:
- microkinetic nanoreactor-level theory
- detailed balance Monte Carlo simulations
- molecular-level synthesis and atomic-level materials characterization
- nanoreactor-level characterization of photophysical and photochemical properties

CROSS-CUTTING Correlative Microscopy Platform | Multiscale Simulation Network | Nanomaterials Synthesis & Characterization | Microscopy Bridge Program

Chemical Selectivity (C)
(Esposito, Hurst, Ogitsu, Salako)

Goals: *(i)* understand in atomistic detail the function of encapsulated molecular reaction centers; *(ii)* codesign spatially distinct interphases for species-specific permeabilities and stabilities at low flux

Methods and Approaches:
- atomistic first principles theory
- molecular dynamics simulations
- molecular-level spatial control of multicomponent surface coatings
- nanoreactor-level characterization of electrical and electrochemical properties

Ensemble Effects (D)
(Bala Chandran, Esposito, Hu, Sambur, Talin)

Goals: *(i)* experimentally validate interparticle interactions mediated by light and matter; *(ii)* guided by simulations bridging microenvironments, program desired physicochemical properties

Methods and Approaches:
- multiscale continuum-level models of coupled physicochemical processes
- characterization of the mutual influence of several proximal nanoreactors
- ensemble-level characterization to identify coupled micron-to-millimeter-scale behavior

Fig. 1 | **EPN**'s 4 multifaceted thrusts that are revealing design rules for ensembles of solar water splitting nanoreactors.

A key hypothesis within **EPN** is that molecularly-precise multicomponent interphases can be synthesized with nanoscale spatial control to independently control reaction selectivity for each of electrons, holes, chemical reactants, and chemical products. Building on knowledge gained from planar model systems, synthesized nanoreactors are being studied in isolation and as ensembles containing several-to-millions of nanoreactors. Outcomes from simulations and experiments are informing codesign strategies for the four interacting microenvironments critical to **EPN**: (1) semiconducting solid phases; (2) multicomponent electrocatalytic interphases; (3) intervening aqueous liquid phases between adjacent nanoreactors; and (4) collective blackbody and solar radiation fields. Through coupled control of these microenvironments, **EPN** is uncovering pathways to achieve high quantum yields and energy efficiencies for all elementary steps, from transport of incident solar photons to formation of chemical products. To better understand how ensemble behaviors arise from outcomes of elementary steps, **EPN** is isolating and characterizing the effectiveness of individual steps, as well as observed reactivity from several coupled steps.

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 are being 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 is being 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 are being interpreted using a multiscale simulation network that 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 are being 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. This will motivate integrated efforts and close collaborations in nanoreactor development that will be achieved using bottom-up nanomaterials synthesis and characterization. Furthermore, knowledge gained from **EPN** is providing 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 is diverse, including AANAPISI, ANNH, HSI, NASNTI, and PBI minority serving representation 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** has developed a Microscopy Bridge Program aimed at training the next generation of scientists and engineers interested in microscopy and solar fuels, while strengthening the STEM pipeline between PUIs and R1 institutions through mutually beneficial research partnerships. Institutional proximity helps facilitate two-way student and PI exchanges.

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

Contact: Shane Ardo, Director, ardo@uci.edu
949-824-3796, https://photosynthesis.uci.edu

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 be 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	Joel Haber
Case Western Reserve University	Burcu Gurkan
Elizabeth City State University	Bijandra Kumar
Lawrence Livermore National Laboratory	Christopher Hahn (Deputy Director)
Oak Ridge National Laboratory	Robert Sacci, Gabriel Veith
Pennsylvania State University	Michael Janik
University of California, Davis	Jesus Velazquez
University of California, Los Angeles	Anastassia Alexandrova, Carlos Morales-Guio
University of Central Arkansas	Marsha Massey
University of Colorado, Boulder	Wilson Smith
University of Louisville	Joshua Spurgeon
University of Michigan	Charles McCrory

Contact: Jenny Yang, Director, j.yang@uci.edu
https://carbonsolution.uci.edu/

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.

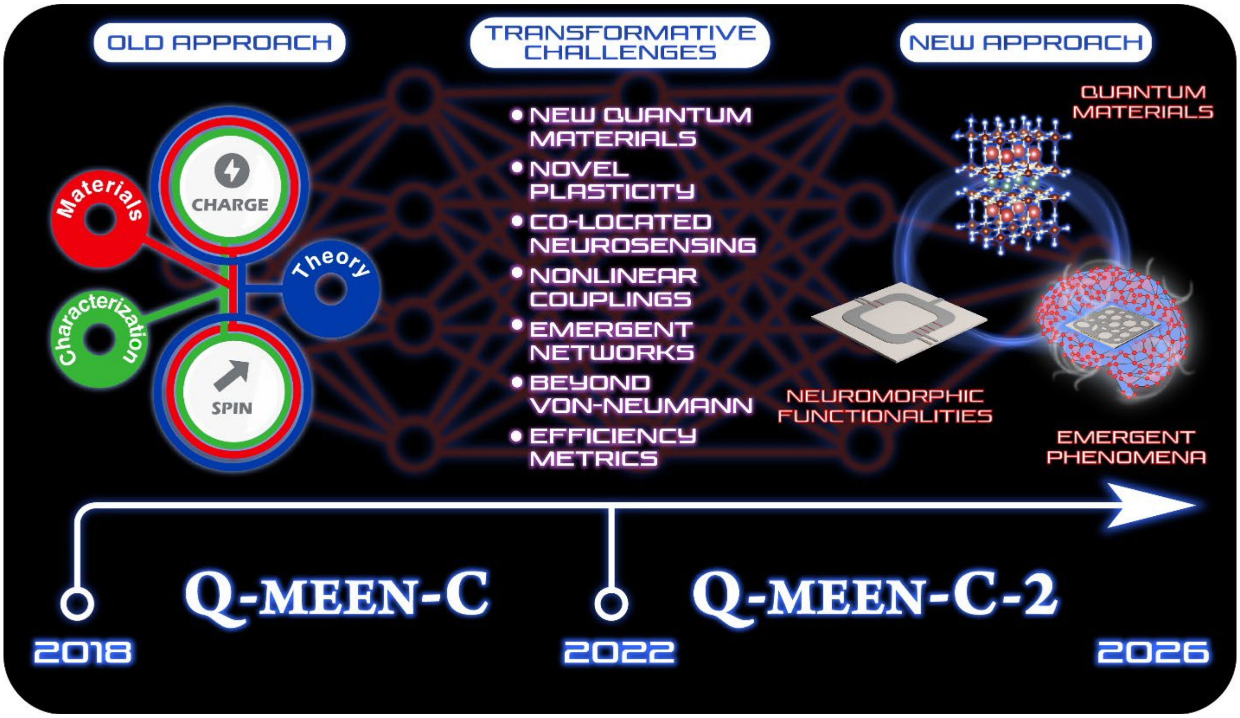


Figure 1: *Based on the accomplishments in the first 4 years with Spin and Charge thrusts, we identify 7 transformative challenges which lead to our newly structured, three-pronged approach.*

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.

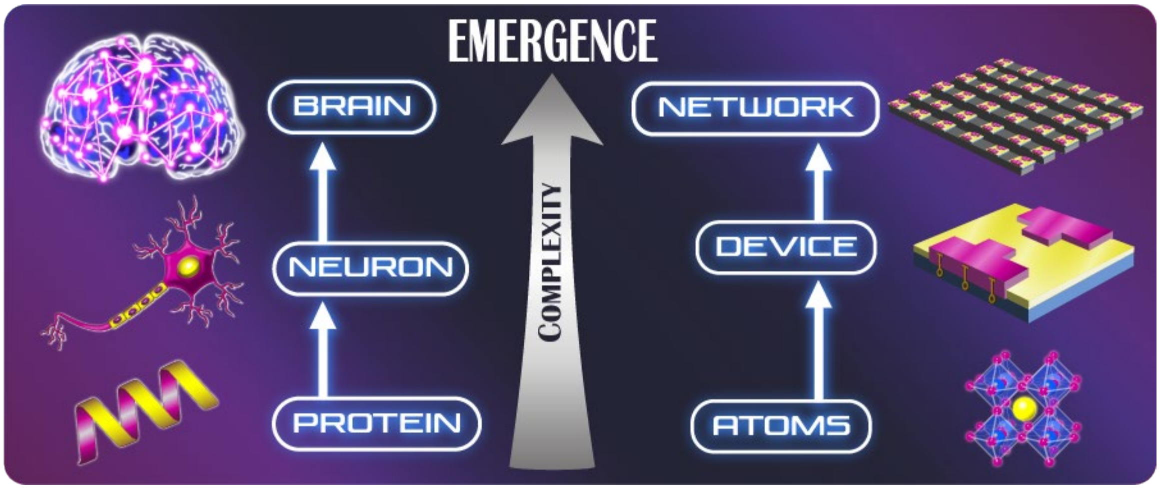


Figure 2: *We mimic the emergent complexity that makes the brain an efficient computer by harnessing the properties of quantum materials, approaching the problem at various length scales.*

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
University of Tennessee, Knoxville	Katie Schumann
Brookhaven National Lab	Yimei Zhu

Contact: Ivan K. Schuller, Director, ischuller@ucsd.edu
858-534-2540, https://qmeenc.ucsd.edu/

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 is being 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 targets 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 are being achieved through interactive and iterative efforts of synergistic computational and experimental techniques that leverage our unique, multi-disciplinary team. These themes 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, is working to 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 drives 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 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 requires selective, low-energy processes for generating higher-order chemical feedstocks from biogenic sources or recycled waste. We also examine reaction mechanisms for C–C bond formation, such as the coupling of CO, carboxylates, or alcohols, which also 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)	
University of Chicago	Laura Gagliardi (Director); John Anderson; Chibueze Amanchukwu; Andrew Ferguson; Ian Foster; Juan de Pablo; Anna Wuttig
Argonne National Laboratory	Massimiliano Delferro
Northwestern University	Omar Farha; Joseph Hupp (Deputy Director); Justin Notestein
Ohio State University	Rachel Getman
Pacific Northwest National Laboratory	Johannes Lercher
Stony Brook University	Karena Chapman
University of Illinois at Chicago	Ksenija Glusac
University of Minnesota	Matthew Neurock; Donald Truhlar

Contact: Mónica Lugo, Managing Director, lugom@uchicago.edu
312-320-3751, cd4dc.center.uchicago.edu

Center for Plastics Innovation (CPI)
EFRC Director: LaShanda Korley
Lead Institution: University of Delaware
Class: 2020 – 2028

***Mission Statement**: To develop energy-efficient, selective, and tolerant chemo- and bio-catalytic and synthetic pathways to valorize diverse plastics waste streams and dramatically increase circularity.*

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 Florida; the University of North Carolina at Chapel Hill; the University of Pennsylvania; and Oak Ridge National Laboratory (ORNL) to shift the landscape of the plastics life cycle from linear (*i.e.,* "make, use, and dispose") to circular (*i.e.,* "reuse, redesign, and repair") *via* a new frontier of valorization and synthetic approaches. We build upon our previously developed foundational, mechanistic knowledge and leverage a cross-cutting core to advance sustainability in the plastics lifecycle. Our research is organized into three distinct, yet synergistic thrusts (**Figure 1**), each targeting a key challenge in plastics circularity. *Thrust 1 targets deconstruction strategies for existing, commodity, non-polyolefin waste* [*e.g.*, poly(methyl methacrylate) (PMMA) and polyamides (PAs)] by developing novel, efficient, robust, and scalable valorization approaches. *Thrust 2* facilitates the *deconstruction of complex, heterogeneous, plastics waste streams* including consumer products that contain additives or multiple layers of diverse polymers. *Thrust 3* moves beyond existing waste streams to design new*, inherently circular polymers* benchmarked against commodity plastics.

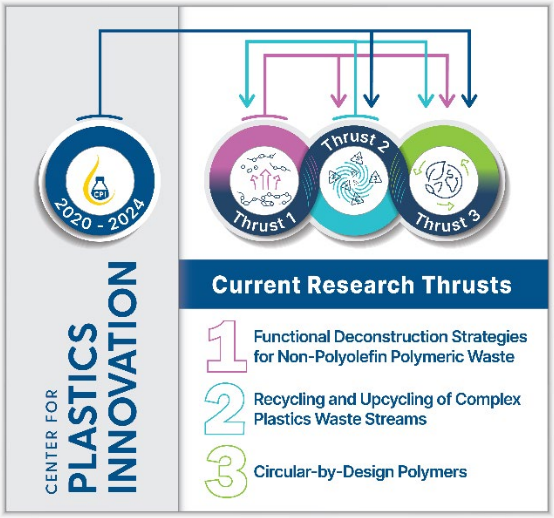


Figure 1. *CPI research thrusts.*

Background and Knowledge Gaps: Plastics manufacturing is a cornerstone of innovation across multiple sectors, including infrastructure, food and medical packaging, aerospace, and energy conversion and storage, enabled by the durability, versatility, low thermal conductivity, high strength-to-weight ratio, and barrier properties of polymers. These technological advances are driving increased demand for plastics production (~1,230 Mt/year by 2060 - OECD *Global Plastics Outlook*; 2022), while introducing higher levels of complexity (*e.g.,* additives, fillers, blends, multilayers) within the manufacturing cycle. There is a pressing need for *fundamental approaches that address the entire plastics life cycle*, from polymers designed for circularity to end-of-life treatment (*e.g.,* deconstruction/reprocessing) of complex polymer waste streams, to mitigate environmental and health impacts worldwide. It is imperative that scientific breakthroughs are realized both downstream (*i.e.,* complex, multicomponent, plastics waste streams after use) and upstream (*i.e.,* pre-manufacture, readily-recyclable, polymeric feedstocks). To unlock these innovations, scientific challenges related to bond energetics, low thermal conductivity, high viscosity, and multicomponent formulations must be recognized and addressed in an interdisciplinary framework. *CPI* is uniquely positioned to deliver *disruptive*, *transformative*, and *robust* solutions to realize sustainable plastics manufacturing, integrating a systems-based and data-driven approach with molecular-level understanding; selective and efficient catalytic pathways; advanced synthetic methodologies; high-throughput screening; and comprehensive, multiscale experimental and computational characterization.

Impact: Transforming the landscape of plastics manufacturing towards circularity requires fundamental strategies that address the complexity of plastics waste streams, emphasize product performance, and enable polymer redesign. We explore deconstruction routes that synergize chemical and enzymatic approaches to produce building blocks from heteroatomic polymers; define pathways to enable tailored deconstruction of waste streams with compositional complexity; and employ methods to impart selective sites for degradation, all to nucleate a paradigm shift in the plastics life cycle *via* detailed mechanistic understanding. Additionally, we develop cross-cutting platforms and tools to enable plastics circularity that will have long-lasting influence on plastics design, manufacturing, processing, and (re)use. Moreover, the coupling of experiments, modeling, artificial intelligence (AI), polymer chemistry/engineering, and life-cycle assessment (LCA)/technoeconomic analysis (TEA) in *CPI* will define new frontiers in trainee development with direct impacts on sustainable manufacturing. Furthermore, these research innovations will provide a platform to foster *belonging*, *leadership*, *access*, and *inclusivity* through community-building activities, active mentoring and outreach, and professional development programming.

Overarching Goals and Objectives: *CPI* addresses the plastics life cycle through three **overarching research goals** in three synergistic research thrusts: 1) develop novel synthetic approaches to valorize major plastics and polymer systems [including thermosets and composites], beyond polyolefins; 2) design strategies to tackle the complex plastics mixtures that are prevalent in 'real waste streams' and mitigate the effects of additives in polymer formulations; and 3) create readily-recyclable polymers with targeted properties as circular alternatives to current materials. These goals will be achieved *via* six targeted **objectives**: 1) design a suite of valorization approaches to enable *low-energy, controlled deconstruction* of non-PO waste streams to monomer and other functional, small-molecule products; 2) develop chemical recycling design principles to address the *compositional complexity of 'real' polymer waste*; 3) design *hierarchical, multiscale catalysts* with tailored and selective catalyst-polymer interactions to facilitate deconstruction of complex and varied waste streams; 4) develop *advanced computational models, theoretical frameworks, and characterization methods* to enable prediction and analysis of deconstruction products, polymer properties, and macromolecular transformations; 5) generate new *structure-property paradigms for circular-by-design polymers via* innovative synthetic schemes with a focus on property enhancement; and 6) employ *systems analyses* to inform strategic decision making to enable circularity.

Center for Plastics Innovation (CPI)	
University of Delaware	LaShanda Korley (Director), Thomas Epps, III (Deputy Director), Michael Berg (Science & Technology Director), Hui Fang, Marianthi Ierapetritou, Aditya Kunjapur, Dongxia Liu, Raul Lobo, Eleftherios Papoutsakis, Dion Vlachos, Mary Watson
Oak Ridge National Laboratory	Josh Michener
University of Chicago	Juan de Pablo, Stuart Rowan
University of Florida	Brent Sumerlin
University of North Carolina at Chapel Hill	Frank Leibfarth
University of Pennsylvania	Daeyeon Lee, John Vohs

Contact: LaShanda Korley, Director, lkorley@udel.edu
302-831-0937; https://cpi.udel.edu

Center for Molecular Magnetic Quantum Materials (M2QM)
EFRC Director: Xiaoguang Zhang
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^2QM)** is to discover, develop, and deliver the pivotal materials physics and chemistry knowledge of molecular magnetic quantum materials essential for quantum information technologies. M^2QM 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^2QM 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.

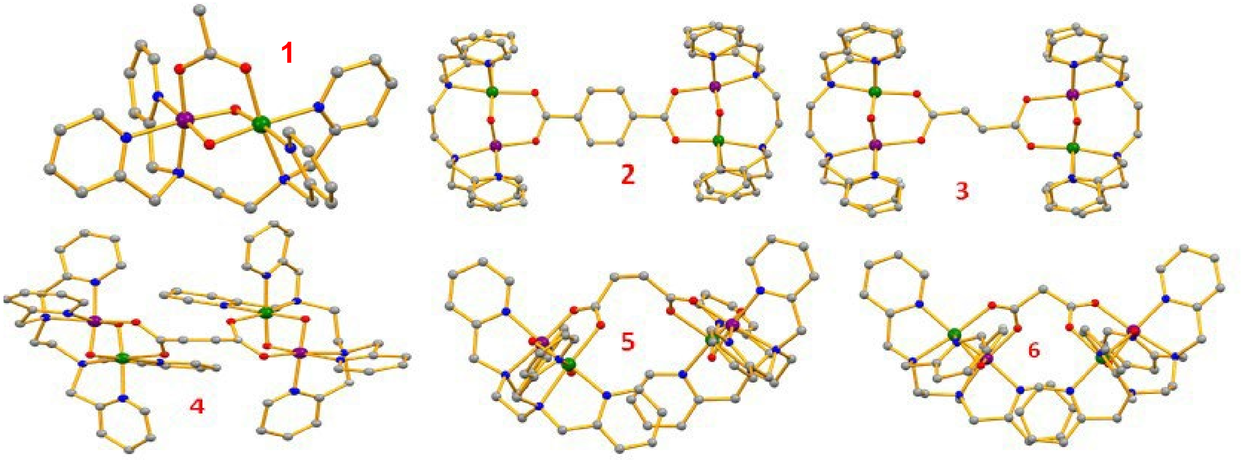


Fig.1 The [Mn_2] magnetic molecule with S=1/2 {1} and its conversion into dimers 2-6 by covalent linkage with a variety of dicarboxylates.

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^2QM 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.

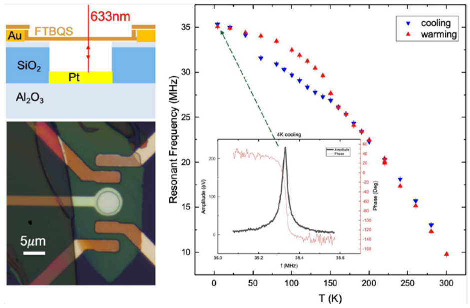


Fig. 2. Spin crossover measurement of 2D molecular film Fe(tBu₂qsal)₂ using mechanical resonator.

We envisage three fundamental directions for the second M^2QM 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, measurements of spin crossover in an $Fe(tBu_2qsal)_2$ thin film demonstrating hysteresis between cooling and heating.

M^2QM'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^2QM'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^2QM)	
University of Florida	Xiaoguang Zhang (Director), Neil Sullivan (Associate Director), Chunjing Jia, George Christou, Arthur Hebard, Richard Hennig, John Stanton, Samuel Trickey, Xiao-Xiao Zhang
Florida State University	Stephen Hill, Mike Shatruk
University of Texas at El Paso	Mark Pederson
Los Alamos National Laboratory	Vivien Zapf
California Institute of Technology	Garnet Chan
Northeastern University	Hai-Ping Cheng (Scientific Director)

Contact: Xiaoguang Zhang, Director, m2qm.efrc@phys.ufl.edu, xgz@ufl.edu
352-392-6256, www.efrc.ufl.edu

Quantum Sensing and Quantum Materials (QSQM)

EFRC Director: Peter Abbamonte
Lead Institution: University of Illinois
Class: 2020 – 2026

***Mission Statement**: To apply advanced scattering and scanning probe spectroscopy techniques to study charge dynamics in quantum materials.*

The modern understanding of quantum materials is based on measuring a few fundamental Green's functions, which describe how excitations propagate in many-body systems. These include the one-electron Green's function, spin susceptibility, and charge susceptibility. While the first two are well measured by techniques like angle-resolved photoemission (ARPES), scanning tunneling microscopy (STM), and inelastic neutron scattering (INS), no equivalent probe has existed for the charge response. The mission of the QSQM is to measure this charge response in various quantum materials, with meV energy resolution and either spatial or momentum resolution. We aim to focus on the following phenomena:

Strange metals. First discovered in copper-oxide superconductors, the strange metal phase appears to be a universal state of matter found in a wide range of materials, including ruthenium oxides, heavy fermion systems, organic molecular crystals, and twisted van der Waals materials. Its defining feature is a universal scattering rate governed solely by fundamental constants, known as Planckian dissipation. We are using momentum-resolved electron energy-loss spectroscopy (M-EELS), along with other techniques, to explore the possibility of conformal symmetry in these materials, which would suggest a quantum critical and universal phase.

Altermagnets. Originally believed to be a type of antiferromagnet, altermagnets are now considered a distinct class of materials characterized by unique symmetry. The QSQM is investigating the predicted paramagnon polaron excitation, a new collective mode thought to be exclusive to the altermagnetic state.

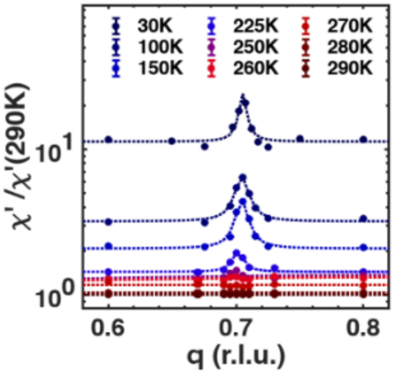


Figure 1 – Susceptibility divergence near the CDW ordering transition in $ErTe_3$. Predicted originally in the 1970's, this effect was measured for the first time this year with momentum-resolved EELS.

Charge density wave materials. Although widely studied, the charge density wave (CDW) instability remains poorly understood. A CDW resembles a superconductor, but breaks a spatial symmetry rather than $U(1)$ gauge symmetry. The CDW is believed to result from a divergence in charge susceptibility at nonzero momentum, though this prediction has never been tested. The QSQM is investigating the behavior of the charge susceptibility near CDW transitions (Fig. 1), focusing on selenide and telluride materials, to gain deeper insights into this unique phase of matter.

Interacting topological phases. The first three-dimensional topological materials identified were topological insulators and Weyl semimetals, where topology is reflected in the single-particle energy band structure and surface states. Researchers are now exploring how topology manifests in strongly interacting many-body systems, where it appears that the relevant excitations are collective modes. The QSQM is employing M-EELS, scanning microwave impedance microscopy (MIM), and scanning single-electron transistor (SET) measurements to investigate collective modes in a variety of interacting topological materials. These include noncentrosymmetric materials with topological phonon band structures, ordered ferroelectrics displaying skyrmion and vortex phases, and two-dimensional fractional Chern insulators.

Quantum Fisher information. Quantum entanglement is a fundamental property of a many-body system, but measuring it experimentally has proven challenging. Recently, it was suggested that the charge response can serve as a measure of quantum Fisher information (QFI) in a many-body system, reflecting both wave function geometry and the density of entanglement. The QSQM is utilizing momentum-resolved scattering techniques to quantify the QFI in quantum materials. We have recently demonstrated that the simple insulator LiF adheres to theoretically predicted bounds, and we are now extending these measurements to more exotic materials, such as strange metals.

Technique development. In addition to conducting pioneering measurements on materials, the QSQM is developing the next generation of instruments that will serve as platforms for future experiments. We are implementing new strategies to enhance the accuracy and speed of momentum-resolved electron energy-loss spectroscopy (M-EELS) measurements. One approach involves using ARPES-type hemispherical analyzers, which enable parallel momentum and energy readout. This is being achieved by integrating a small Scienta SES-100 analyzer with an Ibach-type high-resolution EELS spectrometer and a eucentric sample goniometer. Simultaneously, we are exploring time-of-flight techniques using pulsed electron beams, which will allow for fully three-dimensional measurements, simultaneously measuring energy and two momentum directions in parallel.

Quantum Sensing and Quantum Materials (QSQM)	
University of Illinois, Urbana-Champaign	Peter Abbamonte (Director), Philip Phillips, Tai Chiang
SLAC National Accelerator Laboratory	Ben Feldman, Zhi-Xun Shen

Contact: Peter Abbamonte, Director, abbamont@illinois.edu
217-244-4861, https://iquist.illinois.edu/programs/qsqm

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 addresses 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 seeks 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.

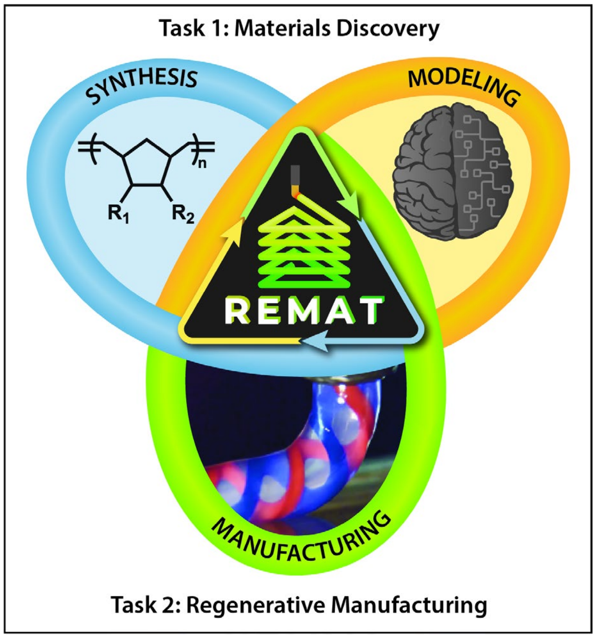


Fig. 1: Organization of the EFRC for Regenerative Energy-Efficient Manufacturing of Thermoset Polymeric Materials (REMAT).

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 for sustainable manufacture of thermoset structures. REMAT's four-year goals and outcomes are distilled into two Level 1 (L1) Milestones:

- ***L1.1*** Develop materials, methods, and models to achieve regenerative thermoset polymers with generation-invariant thermomechanical properties over three lifecycles.
- ***L1.2*** Develop thermo- and photochemical reaction-diffusion processes for regenerative thermoset polymers with fabrication rates, energy consumption, and materials performance that equal or exceed state-of-the-art additive manufacturing methods.

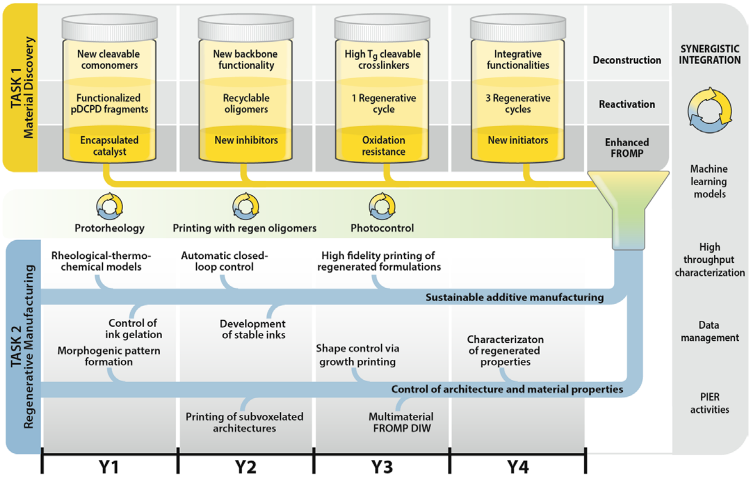


Fig. 2: REMAT roadmap for current and future research.

These goals - the Level 1 Milestones - direct our bottom-up approach to achieving breakthroughs in regenerative thermoset materials. To achieve these goals, REMAT has put in place a highly integrated Task structure around the core competencies of synthesis, modeling, characterization, and processing (Fig. 2). **Task 1, *Materials Discovery for Regenerative Lifecycles (Materials Discovery)***, focuses on highly integrated synthesis, characterization, and modeling to develop machine learning (ML) models that enable predictable formulation of resins with a programmed end of life strategy, the ability to cure efficiently via frontal polymerization and deconstruct and regenerate over multiple cycles, while maintaining properties and performance. **Task 2, *Circular Additive and Morphogenic Manufacturing (Regenerative Manufacturing)***, focuses on sustainable and precise manufacturing of the regenerative resin formulations discovered in Task 1 and control of resulting architecture and material properties. The research flow and subtasks are summarized in Fig. 2. REMAT research progress to date has been greatly accelerated through several synergistic interactions that include activities to promote inclusive and equitable research (PIER). Cross-cutting research interactions have also been enabled by early implementation of cloud accessible, ML ready data management via Clowder, and the development of a unique HTP polymer formulation and characterization facility.

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, Sam Leguizamon
Stanford University	Yan Xia
University of Utah	Matthew Sigman

Contact: Nancy Sottos, Director, n-sottos@illinois.edu
217-333-1041, https://remat.illinois.edu/

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.

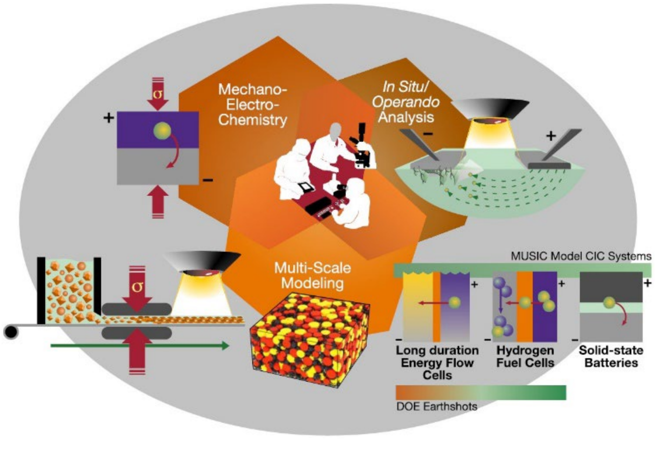


Figure 1: MUSIC will expand the field of mechano-electro-chemistry to accelerate progress toward DOE's Earthshots.

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.

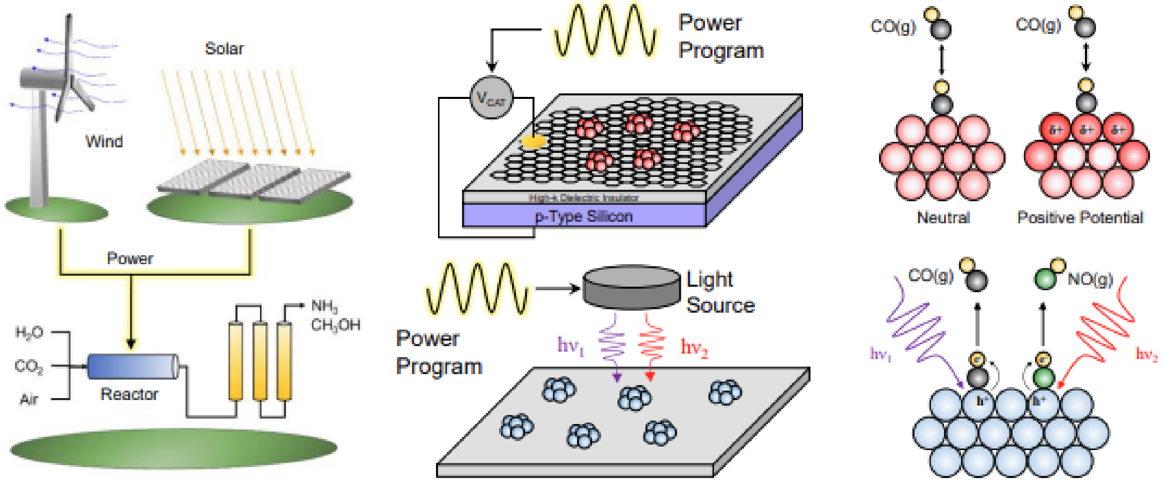
Mechano-Chemical Understanding of Solid Ion Conductors (MUSIC)	
University of Michigan	Jeff Sakamoto, Neil Dasgupta, Bart Bartlett, David Kwabi, Katsuyo Thornton, Michael Thouless
Massachusetts Institute of Technology	Yet-Ming Chiang, Bilge Yildiz
Northwestern University	Sossina Haile
University of Texas, 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
University of California Santa Barbara	Jeff Sakamoto, Raphaële Clément

Contact: Jeff Sakamoto, Director, jeffsaka@umich.edu , sakamoto@ucsb.edu
(734) 763-2219, https://musicefrc.engin.umich.edu/

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

Mission Statement: *To transform how catalysts control energy, and to 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), Dan Frisbie, Aditya Bhan, Bharat Jalan, Matt Neurock
University of California Santa Barbara	Susannah Scott (Deputy Director), Phil Christopher, Mike Gordon
University of Houston	Omar Abdelrahman, Lars Grabow
Ohio State	Rachel Getman
Massachusetts Institute of Technology	Yuriy Roman
University of Michigan	Eranda Nikolla

Contact: Paul Dauenhauer, Director, cpec@umn.edu
612.624.0159, https://cpec.umn.edu

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 that will lead to realizing the potential for the large-scale subsurface storage of CO_2 via mineralization.*

A promising strategy to reduce anthropogenic CO_2 is to permanently mineralize carbon in mafic and ultramafic geologic reservoirs. These rock masses are advantageous due to their prevalence in the earth's subsurface and their ability to rapidly store CO_2 through mineralization. Recent successful pilot-scale mineral carbon storage projects in mafic rock, including CarbFix and CarbFix2 in Iceland and the Wallula basalt sequestration site in Washington State, have demonstrated storage via mineralization on a time scale of a few years. However, storing the levels of CO_2 needed to address the present climate crisis requires (i) a significant up-scaling of these operations and (ii) the ability to predict the impact of long-term, large-scale CO_2 mineralization on the geologic reservoir.

For mafic and ultramafic rocks such as basalt and peridotite, efficient mineralization of the rock mass requires the existence or development of a penetrating fracture network that accommodates the flow and reaction of CO_2-bearing fluids. This involves optimizing fully coupled thermal, hydrological, mechanical, and chemical processes that can sustain flow for long-term carbon mineralization. Complex feedbacks exist among fracture propagation, fluid flow, dissolution, precipitation, and fracture closure, including phenomena such as passivation of mineral surfaces that reduce reactive surface area, carbonate precipitation that can clog pores and fractures, and reaction-driven cracking. These questions require a coupled understanding of fluid flow and transport in fractured media and the chemical and mechanical processes that occur during carbon mineralization. Such a coupled understanding does not currently exist, since the fields of fracture mechanics, fluid flow in fractures, and geochemistry are often studied separately for other subsurface applications such as hydrocarbon extraction, nuclear waste storage, or traditional carbon sequestration where caprocks and seals are used to prevent CO_2 leakage. However, carbon mineralization requires a coupled understanding of reaction rates, feedback between geomechanics and geochemical reactions, and the influence of flow and transport. By expanding the frontiers of interdisciplinary research, GMCS aims to evaluate, for a given CO_2 storage operation within a given reservoir, the evolution of the amount of carbon $\mathrm{M(t)}$ mineralized:

$$M(t) = \int_V m(\boldsymbol{x}, t) dV(\boldsymbol{x}), \tag{1}$$

where V is the volume of the rock mass and $m(\boldsymbol{x}, t)$ is the mineral carbon density (mass of carbon stored per unit volume of rock). All GMCS efforts are anchored by equation (1), with the aim to determine how different mechanisms affect the amount of CO_2 mineralized with time so that the operation can be optimized and up-scaled.

The research GMCS is pursuing aims to fill the knowledge gaps that are the result of this highly coupled fracture mechanics, fluid flow, and geochemical problem. Our research revolves around (i) unlocking the mechanisms behind reaction-driven cracking and fracture, (ii) understanding dissolution and precipitation regimes and their effect on fracture flow, and ultimately combining this knowledge with (iii) continuum and discrete modeling of fracture networks to determine the rate and mass of CO_2 mineralized. The study of these processes is not restricted to a single research thrust and a full understanding is not contained within a single knowledge domain. Indeed, maximizing the success of our research thrusts involve a subtle combination and coupling of the disciplines of geomechanics, geochemistry, porous media flow, and sensing technology. Thus, the research is organized around three thrusts (Fig. 1):

1. Reaction-driven cracking and fracture: elucidate the coupled chemo-hydro-mechanical processes leading to the formation of a microcrack network in the rock matrix.
2. Dissolution and precipitation regimes: understand how mineral carbonation is affected by reactive transport and the interaction between fracture flow and the surrounding rock matrix.
3. Continuum and discrete modeling of fracture networks: develop capabilities to predict and monitor capacity of a mafic or ultramafic reservoir for carbon storage and design optimal solutions for reservoir stimulation.

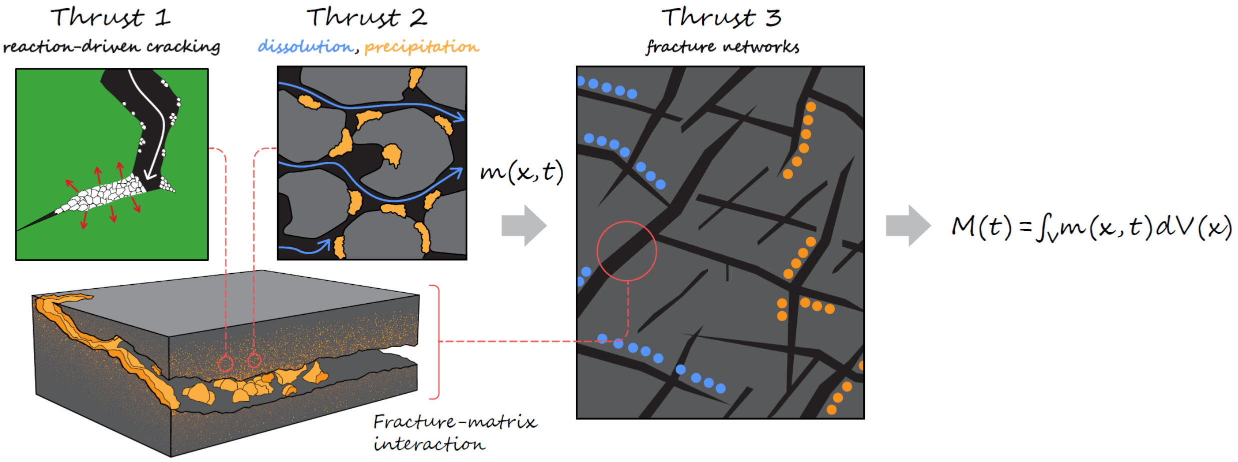


Figure 1. The three thrusts target different aspects of mineralization and share the common goal: the amount of mineralized carbon M(t) will ultimately emerge as a result of competition among the underpinning physical and chemical processes in mafic and ultramafic rocks.

GMCS has launched numerous interdisciplinary projects crucial to the advancement of carbon mineralization. Further, to understand the current state of the science, knowledge gaps, challenges and opportunities, the GMCS team authored a comprehensive review: "Carbon Mineralization in Mafic and Ultramafic Rocks," *Reviews in Geophysics,* submitted. The review covers field studies, laboratory experiments, and numerical simulation, emphasizing that a multidisciplinary approach is required to advance our understanding of what mechanisms control *in situ* carbon mineralization.

Geo-processes in Mineral Carbon Storage (GMCS)	
University of Minnesota	Emmanuel Detournay, Bojan Guzina, Lars Hansen, Peter Kang, Joseph Labuz, Jia-Liang Le, Sonia Mogilevskaya, Vaughan Voller
Georgia Institute of Technology	Haiying Huang
Los Alamos National Laboratory	Jeffrey Hyman, Qinjun Kang, Chelsea Neil, Gowri Srinivasan, Hari Viswanathan
Northwestern University	Zdenek Bazant, Giuseppe Buscarnera, John Rudnicki
University of Southampton	Juerg Matter

Contact: Emmanuel Detournay, Director, detou001@umn.edu
612 625 3043, https://gmcs.umn.edu

Center for Hierarchical Waste Form Materials (CHWM)

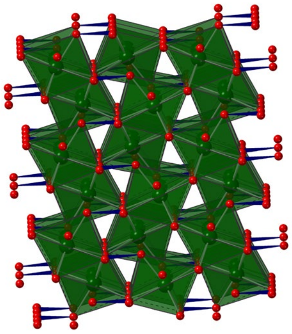
EFRC Director: Hans-Conrad zur Loye
Lead Institution: University of South Carolina
Class: 2016 – 2025

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

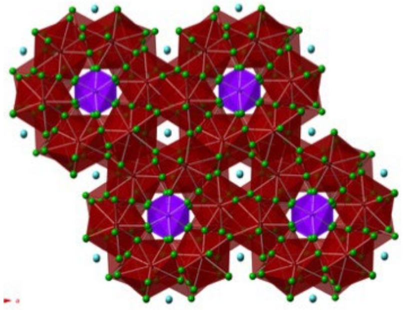
Goals:

The unique predictive and synthetic capabilities developed in the CHWM will be used to study the crystal chemistry of transuranic (TRU) containing extended crystalline structures and their response to radiation damage and radiolytic processes. The Center endeavors to understand the unique chemistry of TRU elements, their influence on the atomic environment affecting the formation of extended structures, their radiation interactions with matter, and how far-from-equilibrium processes can generate metastable phases that are yet quite robust. We hypothesize that an understanding of which types of structural elements favor radiation resiliency of TRU-containing extended structures in waste form materials requires an atomic level understanding together with detailed knowledge about structural changes caused by internal radiation damage. We furthermore hypothesize that the presence of chemically induced anti-site mixing in structures will lead to enhanced self-healing of internal radiation damage.

TRU Containing Oxides and Fluorides:


Structure of $AmBO_3$

We are pursuing oxides and fluorides able to accommodate Np, Pu, Am, and Cm, where we have synthesized and determined the structures of numerous new extended structures, including plutonium (IV) silicates - $K_2PuSi_2O_7$ and $Rb_2PuSi_6O_{15}$; Am containing silicates - $K_3Am(Si_2O_7)$ and $Cs_6Am_2Si_{21}O_{48}$; phosphates - $Na_3Am(PO_4)_2$ and $K_3Am(PO_4)_2$; borates - $Ba_3Am_2(BO_3)_4$ and $AmBO_3$; borate halides - $Ca_5Am(BO_3)_4Cl$; molybdates – $Li_{0.5}Am_{0.5}MoO_4$; and fluorides - $CsAm_2F_7$. Going forward we will pursue Cm containing analogs. Single crystals of non-rad analogs, including some belonging to the structural families discussed above, are being used in radiation damage studies to elucidate their radiation resilience and ability to self-heal.

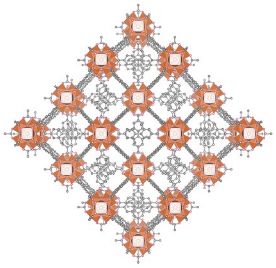

Structure of $Na_3AlPu_6F_{30}$

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-, Cr-, and Ni- substituted hollandites, $(Ba,Cs)_{1.33}(Al/Cr/Ni,Ti)_8O_{16}$, and determined 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 and synchrotron scattering measurements are underway. A parallel SEM/TEM study will correlate local structure features with macroscale determined stability of Cs. Irradiation studies reveal this class of materials has a high tolerance for radiation damage.

Novel Actinide-containing MOFs:

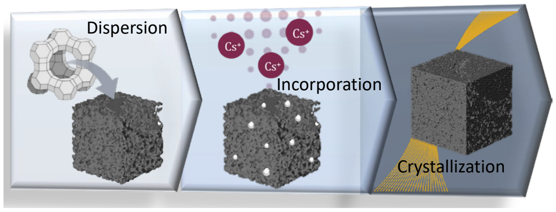
Actinide-MOFs (M = U, Th, and Pu) were prepared via metal node expansion and through the *de novo* synthesis, resulting in the preparation of materials possessing monometallic and heterometallic nodes. Pu-MOFs have been prepared and structurally characterized. We experimentally demonstrated that the 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. Furthermore, we demonstrate that 99% of captured Am cations can be retained at least for several months in the MOF matrix.



Single-crystal structure of Pu-MOF, *Pu-Me_2BPDC-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 determine the thermodynamic 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 forms. 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 Cs-containing 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
Savannah River National Laboratory	Jake Amoroso, David DiPrete

Contact: Hans-Conrad zur Loye, Professor, zurloye@mailbox.sc.edu
803-777-6916, https://chwm.sc.edu

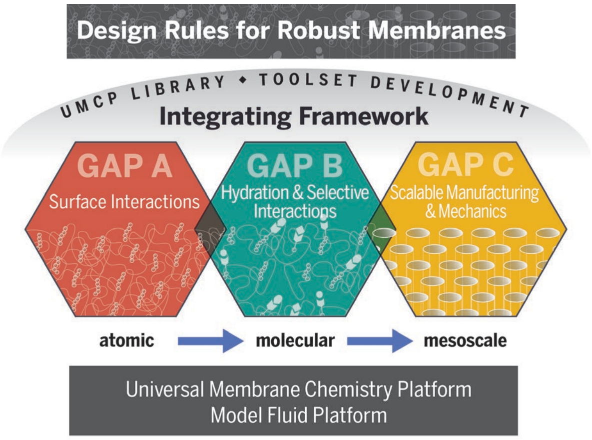
The Center for Materials for Water and Energy Systems (M-WET)

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.

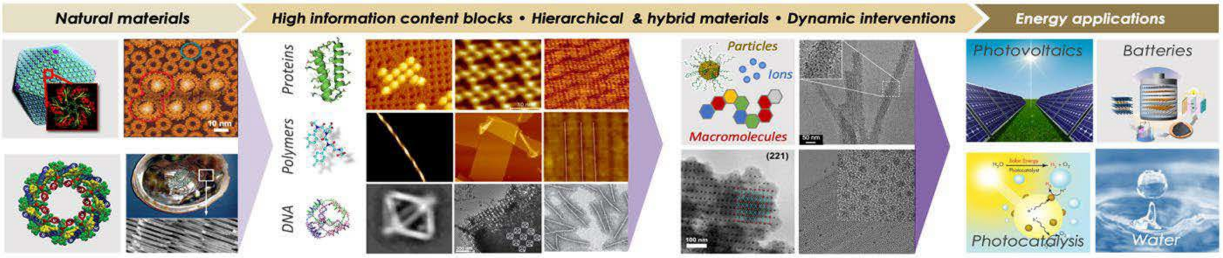
Today, specific interactions (and non-interactions) facilitate solute selectivity and fouling resistance, yet they cannot currently be "designed" *a priori* into membranes. To do so requires fundamental insights into the impact of functional groups on water dynamics near interfaces, an understanding 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)	
The University of Texas at Austin	Benny Freeman (Director), Lynn Katz (Associate Director), Berkin Dortdivanlioglu, Venkat Ganesan, Graeme Henkelman, Manish Kumar, Gabriel Sanoja Lopez, Nathaniel Lynd, Zachariah Page
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 Lab	Ethan Crumlin, Dylan McReynolds, Gregory Su

Contact: Benny Freeman, Director, freeman@austin.utexas.edu
512-232-2803, mwet.utexas.edu

The Center for the Science of Synthesis Across Scales (CSSAS)
EFRC Director: François Baneyx
Lead Institution: University of Washington
Class: 2018 – 2026

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

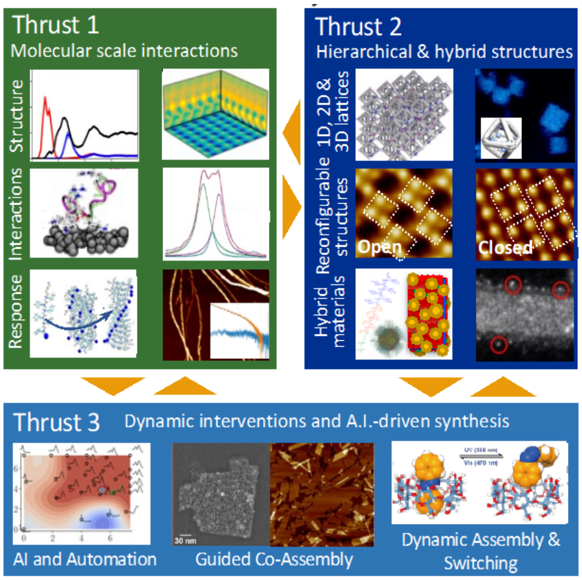


CSSAS: Predictive synthesis of hierarchical and hybrid materials to enable complex energy functions.

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 scientific gaps (SGs) stand between us and that vision:

1. While we have significantly grown the understanding of how building block sequence and chemistry translate into the molecular interactions and assembly dynamics by which order emerges (**SG1**), there are major opportunities to expand this knowledge.
2. While we have succeeded in connecting atomistic descriptions of intermolecular interactions with coarse grained (CG) models of building blocks to bridge the time and length scales required for predicting assembly outcomes of peptides and polymers (**SG2**), we cannot yet fully account for the crucial role of inorganic interfaces, solution conditions, and kinetics.
3. Although we understand many of the principles that underpin macromolecular assembly and inorganic morphogenesis by assembled scaffolds, we are still building an understanding of how the interplay of solvent, electrolytes, disparate building blocks, and dynamic processing conditions control energy landscapes across which assembly proceeds (**SG3**).
4. Finally, our ability to predict metastable states on the pathway to the final ordered state, or to encode a balance of forces that will, by design, create multi-well potentials for out-of-equilibrium switching in response to external stimuli (**SG4**) remains in its infancy.

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, Columbia University, and North Carolina State University, CSSAS will fill these knowledge gaps by tackling three research goals:



CSSAS's thrusts integrate building block synthesis, *in situ* characterization, simulations, and AI to control order, hierarchy, and hybrid states of matter.

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 and AI-driven synthesis, 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)	
University of Washington	François Baneyx (Director), David Baker, Brandi Cossairt (Thrust 2 Lead), David Ginger, Lilo Pozzo (Thrust 3 Lead)
Pacific Northwest National Laboratory	Jim De Yoreo (Deputy Director), Chun-Long Chen, Chris Mundy (Thrust 1 Lead), Wendy Shaw, Shuai Zhang
University of Chicago	Andrew Ferguson
University of California San Diego	Akif Tezcan
University of Tennessee, Knoxville	Sergei Kalinin
Columbia University	Oleg Gang
North Carolina State University	Jim Pfaendtner

Contact: François Baneyx, CSSAS Director, baneyx@uw.edu
206-685-7659, https://www.cssas-efrc.com/

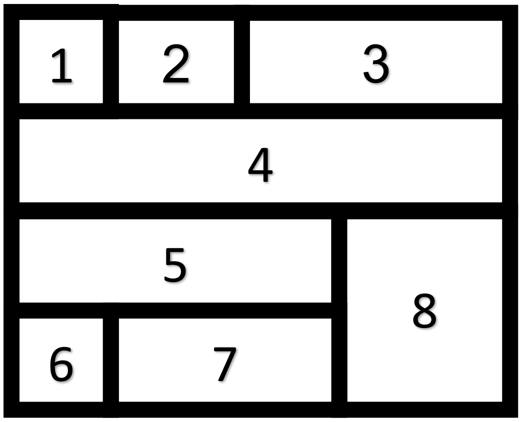
GRAND CHALLENGES INDEX

TRANSFORMATIVE OPPORTUNITIES INDEX

BES REPORTS INDEX

ADDENDUM

FRONT COVER IMAGE



1. Ensembles of Photosynthetic Nanoreactors (EPN) led by Shane Ardo at University of California, Irvine
2. Mechano-Chemical Understanding of Solid Ion Conductors (MUSIC) led by Jeff Sakamoto at University of Michigan
3. A Center for Power Electronics Materials and Manufacturing Exploration (APEX) led by Nancy Haegel at National Renewable Energy Laboratory
4. Quantum Photonic Integrated Design Center (QuPIDC) led by Libai Huang at Purdue University
5. Center for the Advancement of Topological Semimetals (CATS) led by Robert McQueeney at Ames Laboratory
6. The Center for Catalysis in Biomimetic Confinement (CCBC) led by Cheryl Kerfeld at Michigan State University
7. Catalyst Design for Decarbonization Center (CD4DC) led by Laura Gagliardi at University of Chicago
8. Breakthrough Electrolytes for Energy Storage and Systems (BEES2) led by Burcu Gurkan at Case Western Reserve University

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