Resolving the Paradox of Rhizosphere Effect on Soil Carbon Cycle

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The rhizosphere, the interface between plant roots, microbes, and their surrounding soil matrix, is a dynamic and complex system that is critical for the functioning of terrestrial ecosystems. One of the most important functions of the rhizosphere is its role in regulating the cycling of carbon between earth and air. On a global scale, the combined activities of plant roots and soil microorganisms within the rhizosphere release an estimated 3–10 fold more carbon dioxide than the emissions from fossil fuel burning but under the right conditions, soil organic carbon (SOC) can be entrapped in soil aggregates, so it is not released back into the atmosphere. Paradoxically, roots contribute to both stabilization and destabilization of SOC. Rhizosphere processes have the intriguing capacity to both enhance and disrupt the preservation of long-lasting SOC, with an estimated global potential for carbon sequestration as substantial as 5.3 gigatons of carbon dioxide annually. This research addresses the core mission of the Carbon Negative Energy Earthshot by understanding how plant roots affect SOC accumulation and the mechanisms that regulate SOC gain and loss in the rhizosphere by the action of roots, microbes, and soil structure. One possible pathway is root-driven soil aggregate turnover, which encompasses processes such as root penetration, drying-rewetting cycles, and the binding of organic compounds to clay minerals. This pathway plays a significant role in SOC stabilization and destabilization. Another possible pathway is exudate-driven microbial turnover, which involves microbial activities fueled by plant exudate. This pathway influences substrate utilization efficiency and the burial of carbon-containing necromass, both of which have implications for SOC dynamics. The objectives of this research are to quantify carbon processes and understand the rhizosphere pathways by using novel high spatial resolution positron emission tomography (PET) and computed tomography for dynamic data collection of the undisturbed sample volumes both at the root surface and in soil away from the root surface. Traditional static PET imaging yields the time-averaged, spatial distribution of carbon radiotracers, permitting estimates of their accumulation in soil aggregates and other rhizosphere volumes of interest. However, static imaging alone falls short in capturing the dynamic nature of biological processes and cannot explain mechanisms of carbon stabilization. In contrast, dynamic imaging provides both radiotracer distribution and temporal changes of radiotracer as carbon moves between stable and unstable forms. Further, and most importantly, the sequential dynamic PET frames enable highly quantitative techniques for mapping and quantifying radiotracer distribution, transport, metabolism, binding, and more. The kinematic modeling of physiological processes is the key advantage of dynamic imaging of carbon radiotracers. Integrating direct observations with various isotope tracers such as carbon-11 labeled carbon dioxide, carbon-13 labeled carbon dioxide, and carbon-14 labeled carbon dioxide reveal pathways and related rhizospheric mechanisms. Simultaneously quantifying SOC stabilization and destabilization rates in the inter-connected soil matrix and microbial turnover pathways will facilitate studies in previously unattainable ways and offer valuable insights for improving strategies to enhance soil carbon sequestration. Additionally, these findings hold direct relevance for global soil carbon modeling efforts and have potential to resolve the rhizosphere paradox and the well-documented uncertainty and inconsistencies in existing models.

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A Hybrid Framework of Exascale Simulations, Observations and Deep Learning for System-Level Clean Energy Resilience and Risk Reduction

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The DOE Energy Earthshots initiative targets the development of clean-energy systems to disrupt the evolving impact of global climate change. To meet the aggressive clean-energy objectives set by the DOE requires resilience of these systems against natural hazards, without catastrophic failure or unacceptably diminished clean-energy function. Across regions of elevated seismic hazard, this will include resilience against major earthquakes. Hydrogen storage and deep-water floating offshore wind turbines offer just two examples of systems posing unique seismic safety challenges that will need to be addressed. In the case of offshore wind, the two designated tracts located off the California coast are both in regions of very high seismic hazard. Of course safety and resilience must be paramount, and reliable earthquake-resistant design will be a requirement cross-cutting all these new systems.

In this project, we will leverage recent DOE Exascale Computing Project (ECP) developments in high-performance computing and cutting-edge advances in deep learning to establish a capability for predicting the response of energy systems to major earthquakes. Through the synergistic integration of simulation and observational data, this work will bridge the gap between historical statistics-based empirical earthquake models and physics-based large-scale earthquake simulations to build a hybrid earthquake generator that will learn from both simulations and data. A particular focus will be the inclusion of synthetic data from simulations of rare but damaging earthquakes with data from more frequently occurring smaller observed earthquakes that encapsulate the imprint of subsurface geologic structure at fine spatial scale to enhance the prediction of high-frequency ground motions.

The research objectives of the project include the development of deep-learning architectures and algorithms that will assimilate earthquake simulations and ground-motion observations into a hybrid earthquake scenario generator, the development of a schema to test the intensity measures and spatial-variability statistics of the generated earthquake scenarios against published empirical models and simulations outside the learning dataset, the extension of the simulated and hybrid generated spatial coverage offshore, the development of algorithms to add realistic high-frequency distributions to ECP simulators learned from observations, the epistemic uncertainty quantification of the generated ground motions to guide future investments in data harvesting and algorithmic development, and the synthesis of the hybrid generator into a risk-assessment framework for an interconnected clean-energy network, which will enable the DOE to evaluate alternative risk-reduction solutions.

The hybrid earthquake scenario generator will help ensure the resiliency and cost-effectiveness of future energy systems and other regional infrastructure more broadly. Since the machine-learning algorithms will be agnostic to the source and wave-propagation physics of earthquakes, we envision the hybridized framework as a steppingstone to assessing resilience and risk associated with other hazards threatening energy systems, such as hurricanes, floods, wildfires, and combinations thereof.

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Engineering Sporopollenin and its Carbon Supply

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To significantly enhance the capture of carbon in soils, one of the first major challenges is to store it in a form that is stable so that it is not released back into the atmosphere for centuries or millennia. A second major challenge is to capture carbon in quantities that are sufficiently large to achieve a significant reduction in the amount of atmospheric carbon dioxide. A novel way to address these challenges is to directly capture carbon into plant products that are almost 'indestructible' from degradation, in species that are widely cultivated. Sporopollenin – often referred to as the 'plant diamond' – is such a product. Sporopollenin, the outer shell of pollen grains, was an innovation from terrestrial plants to protect pollen from sources of environmental stress. Because of its critical role in plant survival, sporopollenin is produced by a pathway that is highly conserved among different species. It is also distinct from the plant products most often considered for carbon capture and storage (cutin, suberin, and lignin) because of its extreme resistance to degradation – sporopollenin is stable over centuries or more vs. decades or less. Consequently, introducing the production of sporopollenin in the roots of plants can be an opportunity for large-scale, near-permanent capture and storage of carbon in the soil. This potential could be further maximized if applied to widely planted bioenergy or agricultural crops. The goal of this research is to identify the genes that are required to produce sporopollenin in plant roots and release it in the soil. This goal will be achieved using two alternative and complementary approaches. First, a set of genes previously known to be major regulators of sporopollenin synthesis in the developing plant flower will be selected, to be expressed in the roots of poplar trees. In parallel, previously unknown elements that improve the synthesis, transport, and assembly of sporopollenin in poplar roots will be discovered. To test the effectiveness of these approaches, methods to rapidly modify the gene content of poplar roots and evaluate the consequence of these changes in the root structure and composition will be applied. While poplar is being selected as the target species for this study, because the synthesis of sporopollenin is highly conserved among plant species, discoveries made in this research could be applicable to a wide range of biomass and food/feed/biofuel crops such as maize, sorghum, and sugarcane. Finally, the proposed strategy, when deployed at scale, has the potential to remove substantial amounts of carbon from the atmosphere. Considering typical poplar biomass yields (5–10 dry metric tons/ha/yr) and allocation of that biomass belowground (20–25%), engineering roots to contain 5% of sporopollenin could near-permanently store 32–80 kg/ha of carbon in soils annually. Furthermore, it is estimated that engineering the 36-million-hectare U.S. maize crop for 5% sporopollenin content in roots and stover could enable the sequestration of 54 million metric tons of CO2 per year. This is two- to five-fold the current best-practice estimate for annual, long-term carbon sequestration on maize cropland and would significantly increase soil carbon stocks.

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Ka Mana O Ka Lā: Modeling Our Energy Future

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The Department of Energy's (DOE) Energy Earthshots Initiative sets challenging goals to reduce costs of vital clean-energy technologies through innovation and collaboration. This research addresses fundamental applied-mathematics and computer-science issues required to exploit advanced high-performance computing (HPC) modeling for Energy Earthshots challenges. The work will enhance the HPC code PISALE (Pacific Island Structured-adaptive mesh refinement with ALE) for a wide variety of problems aligned with the aggressive Earthshot goals. PISALE is a multiphysics fluid-based suite developed by University of Hawai'i (UH) researchers in collaboration with DOE labs and other universities. In this work, PISALE will be combined and enhanced with MFEM and AMReX, two major software frameworks that have been co-design cornerstones of the Exascale Computing Project, to create a new overarching yet practical framework highly applicable to a variety of Earthshots. Earthshot-relevant data will be used to benchmark progress, and the code's open-source availability will enable involvement of Earthshot scientists directly. The research addresses fundamental questions, including an assessment of the fluid-based approach, pinpointing its limitations and identifying scales at which other computational devices such as particles are required to model Earthshot experiments. Additionally, it delves into the complicated task of integrating finite-volume-based solvers and adaptive multiresolution grids with finite-element solvers. This integration is essential for modeling a diverse spectrum of relevant physical processes, encompassing phenomena such as conduction, diffusion, and fluid flows within geothermal systems.

"Ka mana o ka lā" indicates we look to the essence and power of the sun for a clean-energy future. Thus the research targets several Earthshot projects based on environmentally prudent energy sources powered naturally through the sun and earth's energy sources: improved capability of geothermal processes related to the Enhanced Geothermal Shot, leading-edge erosion of wind-turbine blades, and advanced modeling techniques at the SLAC Linear Accelerator Center for investigation and design of new materials as part of the Long Duration Storage Shot. Crosscutting aspects of the proposed work include upgrading PISALE's modeling capability to study heating by short-pulse optical/X-ray lasers and ion beams as well as the associated material response, which can be applied to development of new materials that can work in extreme environments. Experiments can also uncover new ways to develop materials requiring less cost/energy; effective modeling of these experiments drastically reduces time to obtain results. The efficient coupling to MFEM and the addition of particle functionality will better position PISALE to address Earthshot goals and experimental modeling.

The University of Hawai'i (UH), Mānoa, leads this project in collaboration with Lawrence Livermore National Laboratory and Lawrence Berkeley National Laboratory. Because UH is centered geographically in a region of crucial importance to Earthshot goals, it is critical to engage UH students and the community in these objectives. This project builds an important partnership between UH and major DOE laboratories focusing on the ambitious Earthshot program.

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Proton and Ion Management in Bipolar-Membrane-Based Electrochemical Systems

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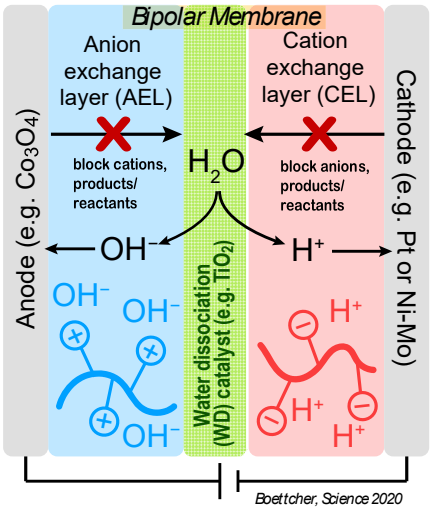
This Science Foundations for Energy Earthshots team will undertake a three-year effort to investigate the fundamental reactions of water and the transport of ions in bipolar membranes and related electrochemical systems. Understanding the generation of ions and controlling their movement is essential to realizing more efficient and lower cost technologies for renewable energy conversion, energy storage, and separations. Bipolar membranes, originally developed for applications in desalination, have emerged as promising components of electrochemical systems that are pertinent to the goals of the *Hydrogen, Long Duration Storage,* and *Carbon Negative* Energy Earthshots. These include electrolyzers for generating green hydrogen from water – including from impure water sources such as seawater – and liquid fuels from carbon dioxide (CO_2), fuel cells for electricity generation from hydrogen, high-energy-density redox-flow batteries for long-duration electrical energy storage, and pH-swing devices for direct CO_2 capture from the air or ocean. Composite membranes will further enable the design of electrochemical devices with functions that are currently impossible to achieve with conventional monopolar membranes.



Bipolar membranes consist of anion- and cation-exchange layers that sandwich a thin film catalyst for water dissociation to H^+ and OH^- (in reverse bias) and acid-base neutralization (in forward bias). To impact Earthshot-relevant applications, these membranes must operate at high current densities (> 1-2 A/cm^2), where the activity of the interfacial catalysts limits their performance. Research will address this problem by combining experimental electrochemistry theory and modeling on molecular to macroscopic length scales, nanomaterials synthesis, and materials characterization. The specific aims of the project are (1) to attain a molecular-level understanding of the catalyzed water dissociation reaction in bipolar membrane junctions, (2) to elucidate the role of the interfacial electric field and proton-concentration gradients in driving water dissociation catalysis and H^+/OH^- ionic recombination, developing multiple methods by which field-enhanced processes can be measured *in operating devices* and modeled, (3) to physically characterize the molecular structure and dynamics of bipolar membrane junctions, (4) to create, based on this understanding, more effective and durable materials and architectures for bipolar-membrane catalysis, and (5) to invent advanced membranes that enable new kinds of electrochemical energy conversion systems.

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Understanding Thermo-Chemo-Mechanical Transformations in Thermal Energy Storage Materials and Composites

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Globally, 90% of energy is associated with the generation, conversion, and/or utilization of heat across a range of temperatures. Thermal energy storage (TES) technologies are thus critical to meet global decarbonization and climate goals. Compared to electrochemical batteries, TES has the inherent advantage of decoupling energy capacity and power output using low-cost materials, which makes it promising for long duration storage. TES can also store electricity and discharge it as heat at high temperatures, which makes it suitable for coupling with industrial processes. These applications require the use of electro-thermal materials, *i.e.*, materials that possess sufficient electrical conductivity for resistive heating during charging, and sufficient thermal conductivity for extracting (discharging) stored energy on demand. This challenging set of simultaneous property constraints cannot be achieved with a single material and requires the design of composite architectures. Furthermore, while the charge-discharge process is central to thermal storage, it is accompanied by large thermal and concentration gradients that cause and are intrinsically coupled to morphological evolution and mechanical stress generation in these materials. These mesoscale structural transformations influence transport and can result in a loss of reversibility during cycling, which leads to storage capacity degradation. In other words, macroscale thermal energy storage is governed by structural evolution at the mesoscale. A fundamental understanding of the storage and degradation mechanisms in these materials across length and time scales under dynamic thermo-chemo-mechanical extremes is thus necessary. The overall goal of this research is to unravel how material composition/structure and mechanical stress co-evolve to determine meso-to-macroscale transformation pathways in TES materials. To provide a holistic understanding of the underlying mechanisms that cause irreversibility during the dynamic storage process, two classes of solid-state TES materials with high energy densities will be studied: thermochemical materials (TCMs) that store energy in reversible solid-gas reactions and high-temperature materials (HTMs) that can access large temperature differences. This will be accomplished through three research objectives: *(i)* mechanistically understand and probe the dynamic evolution of TES materials under thermal, mechanical, and chemical gradients, *(ii)* elucidate and predict the bidirectional coupling between mechanical stress/damage and transport through computational modeling, and *(iii)* design architected composite materials that undergo minimal chemo-mechanical damage during thermal cycling. Drawing inspiration from recent advances in other solid-state energy storage systems to minimize sluggish kinetics, coupled transport, and uncontrolled phase formation in composite materials, this effort will provide fundamental insight of TES material behavior by advancing the state-of-the-art in instrumentation science and developing predictive computational models to guide the design of composite TES structures. The knowledge gained from these studies will catalyze the development of new multifunctional TES materials that are tailored to maximize storage capacity and minimize degradation. A fundamental understanding of TES is relevant to multiple Energy Earthshots – the Long Duration Storage Shot that aims to reduce the cost of grid-scale energy storage by 90% for systems that deliver 10+ hours of duration, and the Industrial Heat Shot that aims to develop industrial heat decarbonization technologies with at least 85% lower greenhouse gas emissions.

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Biological Routes for Synthesizing the Industrial Platform Chemical, Propylene, from Deconstructed Lignin Waste and Captured Carbon Dioxide Produced during Lignin Valorization into Bio-oil

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Each year the chemical industry invests more energy than anywhere else to make ethylene and propylene chemicals for the manufacturing of plastics, textiles, solvents, building materials, and detergents that enable modern society. This energy burden is due to the large amounts of heat required to transform petroleum hydrocarbons into ethylene and propylene and subsequently purify them. Currently these demands are met through the combustion of fossil fuels, leading to significant greenhouse gas emissions, and thereby necessitating the development of alternate processes with low heat and energy consumption. Lignin, a major component of plant biomass is abundantly available but difficult to utilize to produce platform chemicals. Lignin can be transformed by modest heating through a process called Hydrothermal Liquefaction (HTL) to make renewable transportation biofuels. However, about 15% of the processed lignin ends up as aromatic compounds and 15% as gaseous carbon dioxide that are environmentally harmful waste byproducts of the nascent biofuel industry. In recent years, bacteria like *Rhodospeudomonas palustris* have been discovered that can utilize lignin monomers for growth and thus serve as promising chassis for upscaling lignin monomers and CO_2. In addition, *R. palustris* and related bacteria possess newly discovered enzymes distantly related to nitrogenase that can synthesize essential platform chemicals like ethylene, propylene, propanol and propylamine from ubiquitous organic sulfur compounds. Most notably, *R. palustris* performs these biological transformations anaerobically, allowing safe hydrocarbon production by avoiding oxygen combustion hazards. This project will design and implement in *R. palustris* biosynthetic pathways that convert lignin aromatic acids and CO_2 into volatile organic sulfur compounds that serve as substrates to produce propylene, propanol, and propylamine. Next bioinformatics and gene synthesis technologies will enable mining and screening for protein homologs in the engineered pathways that are highly active and increase product yields. Pathway design for optimal conversion of lignin aromatic acids and CO_2 into hydrocarbons will be guided by advanced physics-based thermo-kinetic modeling of *R. palustris* metabolism, which will provide predictive insights on needed enzyme activities, transcriptional regulation strategies, and elimination of pathway bottlenecks. Synergistically, this project will quantify and understand the mechanism and product distribution of the lignin HTL process to generate both bio-oil for transportation and aromatic acids and CO_2 for *R. palustris* to upscale into hydrocarbon platform chemicals. To address CO_2 capture from lignin HTL and delivery challenges to *R. palustris,* this project will develop and understand the fundamental physical principles of new carbon dioxide removal (CDR) technologies involving CO_2 absorbent-coated ceramic honeycomb microchannels and water-based CO_2 desorption in a manner that minimizes energy requirements. Ultimately, this project will generate new technologies that will 1) maximize plant biomass utilization for bio-oil production by the lignin HTL process, 2) minimize thermal energy inputs to produce ethylene and propylene platform chemicals for the growing bioindustry, and 3) contribute to next-generation CDR technologies for capture and biological conversion of CO_2 into biofuels and biochemicals.

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Unleashing Photosynthesis and Nitrogen Fixation for Carbon Neutral Production of N-Rich Compounds

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Reliance on nitrogen fertilizers continues to grow as food production strives to keep pace with a fast-growing world population. Currently, synthetic NH_3 constitutes the heart of the fertilizer industry. The Haber-Bosch process is the principal source of this NH_3 and has a massive carbon footprint, utilizing 1% of the world's total energy production and accounting for ~1.2% of global annual CO_2 emissions. The chemical reaction in this process is run at 500˚C and generates a molecule of CO_2 as a coproduct for every molecule of NH_3 synthesized, leading to ~2.8 tons of CO_2 emission for every ton of NH_3 produced. Thus, the extensive use of synthetic fertilizers in today's agriculture comes with severe environmental penalties, including a significant increase in the global carbon footprint. An alternative approach relies on biological conversion of nitrogen, a greener process restricted to only a few select groups of microbes. Of these, cyanobacteria can drive the energetically expensive reaction solely with solar power while simultaneously capturing CO_2, reducing the carbon footprint. Using non-model cyanobacteria as a chassis to convert atmospheric nitrogen into valuable N-rich compounds requires significant fundamental research activities. This project addresses basic research challenges to develop N_2-fixing cyanobacteria as cell factories to produce nitrogen-rich compounds. The work focuses on the production of guanidine, ammonia, and urea, three nitrogen rich compounds that can serve as substitutes for synthetic fertilizers. While ammonia is a rich source of nitrogen, the use of urea as a fertilizer is equally attractive and often surpasses the global use of anhydrous ammonia. Guanidine, on the other hand, is a slow-release nitrogen fertilizer. Unlike urea and ammonia which have an uptake efficiency of only ~50% by crop plants, leading to significant wastage, guanidium salts can ensure more controlled and protracted release of nitrogen and better utilization efficiency. The project involves two strains representing two contrasting paradigms that cyanobacteria use to accommodate the mutually antagonistic processes of oxygenic photosynthesis and nitrogen fixation: temporal separation in a unicell and spatial separation in a filament. The project will yield novel enzymes capable of catalyzing the conversion of N_2 into guanidine, ammonia, and urea as well as membrane transporters that will secrete the products out of the cell. Multiomics studies and machine learning tools will unravel the fundamental principles underlying the regulation of carbon and nitrogen fixation in cyanobacteria and their channelization towards the products of interest. This technology, when fully developed, has the potential to replace the use of synthetic fertilizers, and addresses the Carbon Negative and Industrial Heat Energy Earthshot goals of the Department of Energy. The team envisions future deployment of such strains in soil as a source of nitrogen and in ocean fertilization for CO_2 removal. Moreover, the fundamental knowledgebase developed could have a broader scientific impact on carbon neutral biomanufacturing of petrochemical replacement compounds and material. The project team brings together significant interdisciplinary expertise in cyanobacterial systems biology, computational modeling, AI, machine learning and synthetic biology. An important mission of this project is to train students and trainees from underprivileged communities. The project team is committed to fostering principles of diversity, equity, inclusion, and accessibility to create a research environment where all participants matter and belong.

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Learning Reduced Models under Extreme Data Conditions for Design and Rapid Decision-making in Complex Systems

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Today's methods for learning reduced models from data are mostly based on batch training, which means that all data are stored in one place, can be accessed multiple times during training, and are fully informative for learning the dynamics of interest. However, the setting is starkly different in many applications in science and engineering, especially in several of the Energy Earthshots and other DOE/ASCR scenarios that involve complex systems with rich physics phenomena and rely on numerical solvers based on large-scale, distributed computing. There, data are computed in a distributed manner across multiple nodes because high-fidelity numerical solvers are typically parallelized via domain decomposition techniques. Full state data vectors of high-fidelity physics solvers are rarely written to disk but instead must be processed in-situ as they are computed during time stepping; both because of storage limitations and because writing to disk can increase the run time per time step tenfold. Additionally, non-equilibrium, tail, and rare events are typically not seen during simulation runs with standard inputs, and thus data generated without active steering contain little information for learning reduced models that are predictive in such critical regions of interest. We propose to go far beyond this current state of the art by developing mathematical foundations and computational methods that enable (i) actively collecting data that are informative about rare events and (ii) learning reduced models from distributed and streamed data. The proposed methodologies will be integrated in computational processes for simulation, design, and decision-making of the Floating Offshore Wind and Carbon Negative Shot, where we will leverage reduced models for speeding up parts of the overall computational process while relying occasionally on the expensive, high-fidelity physics solvers in a principled way to quantify uncertainties and maintain accuracy guarantees.

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Carbon Dioxide Removal and High-Performance Computing: Planetary Boundaries of Earth Shots

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Natural climate solutions aim to remove carbon dioxide from the atmosphere by leveraging the carbon storage potential of natural ecosystems. Two methods currently being explored entail converting carbon dioxide to bicarbonate through spreading minerals on land (enhanced weathering) or adding alkalinity to the ocean by mineral spreading or removing acid (ocean alkalinity enhancement). If employed at scale, these climate solutions could remove atmospheric carbon dioxide and alleviate ocean acidification. Currently, there is no end-to-end framework to assess the impacts of enhanced weathering or ocean alkalinity enhancement, which are likely to be pursued in parallel. This work will create a modeling framework that connects rainfall, soils, rivers, coastal oceans, and open oceans to fully evaluate the impact of these natural climate solutions on atmospheric carbon dioxide concentrations, ecosystems, and ocean pH.

This research was selected for funding by the Office of Biological and Environmental Research.

Harnessing Electrostatics for the Conversion of Organics, Water and Air:
Driving Redox on Particulate Liquids Earthshot (DROPLETS)

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The **overall objective** of DROPLETS is to explore an unconventional, straightforward, and underutilized approach based on microdroplet-enabled redox reactions towards H_2 production, CO_2 activation, and the synthesis of redox species for long-duration energy storage (LDES). In recent years, it has been established that the scope of chemical reactions in aqueous microdroplets produced in a variety of formats significantly departs from that in bulk solutions. Such microdroplets can dramatically accelerate chemical reactions or accomplish complex chemical transformations with simple experimental setups. Although these experiments have unambiguously demonstrated a new pathway for chemical reactions, a concerted team effort to understand their **fundamental principles** leverages their unique properties towards energy applications and CO_2 conversion. To this point, DROPLETS pursues three objectives: 1) the splitting of water to generate renewable hydrogen and oxygenated precursors, 2) the development of water oxidation-driven processes for the electrosynthesis of molecules for LDES and CO_2 capture, and 3) the exploration of mechano-electric effects in the synthesis of organic molecules and precursors for LDES. DROPLETS strives to achieve simplicity and lower the energy input and reactor cost for complex chemical reactions, a key advantage to simultaneously achieve multiple Energy Earthshots, including the Hydrogen Shot, the Long Duration Storage Shot, and the Carbon Negative Shot. This project also features **efficiency in collaborative science**, the use of advanced characterization techniques, and a forward-looking plan for data management. DROPLETS ensures that the scientific diversity of the project's research is also reflected in the **human diversity** of its trainees and scientists. This is accomplished by means of a promotion of inclusive and equitable research (PIER) plan that trains a diverse team and builds a thriving community with equitable access to scientific resources, including two Hispanic-serving institutions. Continuous training, mobility grants, community building through established partners, an annual retreat with components for safety, ethics, data management, and creativity awards, ensure an overall benefit to America's ability to successfully accomplish the Earthshots.

This research was selected for funding by the Office of Basic Energy Sciences.

Design, Discovery, and Synthesis Science of Porous Frameworks using Fast and Modular Heterophase Assembly

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Porous and crystalline materials, such as Covalent Organic Frameworks (COFs), uniquely combine intrinsic porosity and programmable function via a wealth of chemical functionalities, which critically determine activity and selectivity in electrochemical reactions crucial to meeting the targets of the Hydrogen and Carbon Negative Energy Earthshots. To transcend common practices of growth that limit programmable function, the research will focus on studying vapor-phase synthetic pathways for COFs using scalable molecular layer deposition. Electrode surfaces coated with well-defined COF thin films that contain molecularly precise functional groups will be engineered via vapor-phase synthesis to direct desired electrochemical transformations such as electrochemical CO_2 capture and concentration (e-CCC) and electrochemical hydrogenation/dehydrogenation of liquid organic hydrogen carriers (e-LOHC). The precision that can be achieved through this process provides an opportunity to go beyond "single atom catalysts" (SACs), whose chemical simplicity limits the ability to perform advanced electrochemical transformations selectively and efficiently. Growth of these frameworks from the vapor phase could result in molecular assemblies at fast rates with simplified processing and will accelerate the discovery of new materials with transformational properties.

The core mission of this project is therefore to overcome the limitations of current classes of COFs by understanding, predicting, synthesizing, and controlling their structure and surface chemistry with molecular precision and modularity, as needed to impart transformative electrochemistry for e-CCC and e-LOHC. This synthetic approach can be generalized for other framework materials as well, such as metal-organic and zeolitic imidazolate frameworks. This project will produce impactful publications, methodologies, computer codes, and data for sharing with the scientific research community. A core value will be to actively pursue inclusive excellence within the research environment, harness it to bolster intellectual diversity, and thereby leave a profound and enduring impact on the education and development of high school students, graduate scholars, and early-career researchers.

This research was selected for funding by the Office of Basic Energy Sciences.

Process-Based Experimental and Machine Learning Approaches for Controlling Fracture Network Generation in the Brittle-Ductile Crust

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Several of the DOE's Energy Earthshots (Enhanced Geothermal Systems, Carbon Negative, and Long Duration Energy Storage) will benefit greatly from improving our ability to create fracture networks that control fluid flow through the Earth's crust. The primary way to create new fractures is by injecting fluid into boreholes under controlled pressure or flow rate ("hydrofracture"). These fractures propagate at rates that are determined by a rock's composition, structure, pressure, temperature, and other factors. Relevant to the Earthshot goals, when the rock and fluid are at different temperatures, and/or can undergo chemical interactions, the behavior of cracks will be further affected. Knowing and controlling the characteristics and extent of fracture network growth is currently a highly uncertain art, made even more difficult by the very limited ability to directly observe what is happening deep in the crust. The primary source of information is the acoustic waves emitted from cracks as they propagate, and our understanding of these "sounds" is currently minimal. Machine learning (ML) has enormous potential for improving our ability to identify subtle patterns in these sounds and help people interpret them in a way that will enable much higher levels of control than are currently possible. This project aims to build this potential into practical tools.

This project will develop a new approach to ML design that integrates the laws of thermodynamics, namely a particular branch of the "Thermodynamics of Irreversible Processes" (TIP), into a neural network architecture. This "TIP-informed Neural Network" (TIP-INN) integrates both mechanical data and subtle acoustic wave patterns to construct a thermodynamic representation of the behavior of the fluid-rock system. The model constructed by the TIP-INN will be used to predict the mechanical behavior (such as fracture rate and fluid flow) for any rock-fluid system by ingesting data in real time. In this project, the TIP-INN will be built in conjunction with novel laboratory experiments. These experiments involve producing hydraulic fracture in a range of materials (starting with 3-D printed acrylic, moving to marble and other rock systems) across a range of conditions of temperature and stress. Although only the acoustic signals and mechanical data will be fed into the TIP-INN, high-speed video will also be recorded to better understand the fracture processes associated with different patterns in the acoustic signals. This approach will later be applied to some active geothermal reservoirs including DOE-funded test sites (e.g., FORGE and Newberry Volcano projects), to begin the process of real-world implementation.

While the applications of this ML approach are very broad, the focus will be on processes most relevant to high-temperature crustal conditions necessary for large-scale geothermal heat mining, an emphasis of the Enhanced Geothermal Systems Earthshot. Additional considerations will be directed towards exploring chemical conditions (disequilibrium) relevant to carbon sequestration, lithium and other critical mineral extraction, and subsurface energy storage. When fully developed, this ML approach will be used in real-time, to inform reservoir decision-making for optimal operations, to enable a rapid expansion of these subsurface roles for climate-change mitigation.

This research was selected for funding by the Office of Basic Energy Sciences.

Fundamental Studies of Hydrogen Arc Plasmas for High-Efficiency and Carbon-Free Steelmaking

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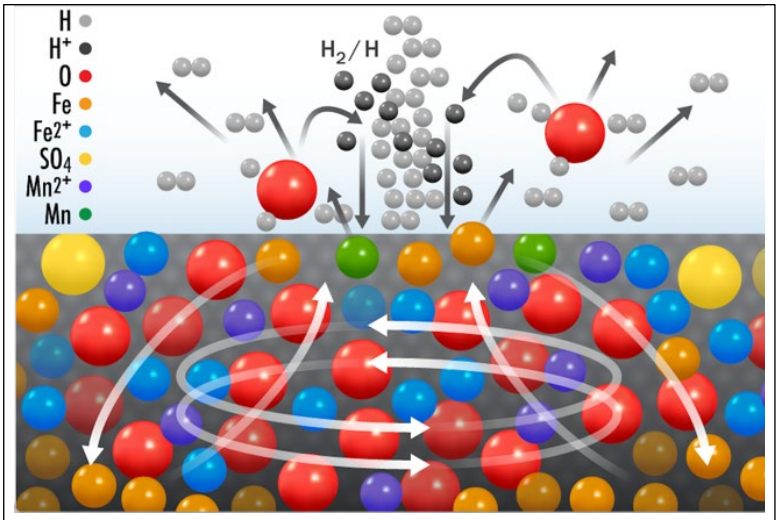
Steelmaking is one of the five most carbon-intensive industries, and accounts for 7% of global emissions of carbon dioxide (CO_2), primarily due to the use of carbon in the process. Hydrogen can be used in place of carbon for producing iron from ore without emitting CO_2, but existing technologies using hydrogen reduction produce *solid* iron containing unwanted impurities (referred to as "gangue") that are difficult to remove using the Electric Arc Furnace (EAF) technology used by most US steel mills today. An alternative process using hydrogen in the plasma state would both improve the rate of reduction and result in a *molten* iron product separated from the gangue. Hydrogen-plasma-smelting-reduction (HPSR) of iron ore — currently only a concept — is therefore a highly promising technology to develop because it would dramatically reduce carbon emissions during steelmaking while also increasing the variety of usable iron ores. This project is a necessary first step to achieving hydrogen-plasma-smelting of iron ore at scale. The work will be carried out by a project team that includes researchers from Arizona State University and the University of Texas at Austin, both Hispanic Serving Institutions, and Navajo Technical University, the largest tribal college in the US, along with the National Renewable Energy Laboratory (NREL). The project will conduct laboratory scale experiments at the extreme conditions, expected in the proposed plasma environment, to gain an in-depth understanding of the basic science behind HPSR. These experiments will then be used to develop theoretical models that will require Exascale computing and advanced machine learning techniques to couple all relevant physics including hydrogen arc physics, melt pool hydrodynamics, phase transitions in metals and oxides, and the kinetics of iron ore reduction. The fundamental science insights and theories from this project will enable the steelmaking community to design and develop technically- and economically viable reactors for carbon-free steelmaking. This project contributes to DOE's climate and economic competitiveness goals related to the carbo- free process heating and metal-oxide reduction which is relevant to the Industrial Heat Shot.



A schematic of the molecular interactions that take place during hydrogen-plasma-smelting-reduction of iron ore.

This research was selected for funding by the Office of Basic Energy Sciences.

Molecular and Atomic EngineeRing of Interfacial Electro-catalytic Environments (MARIE)

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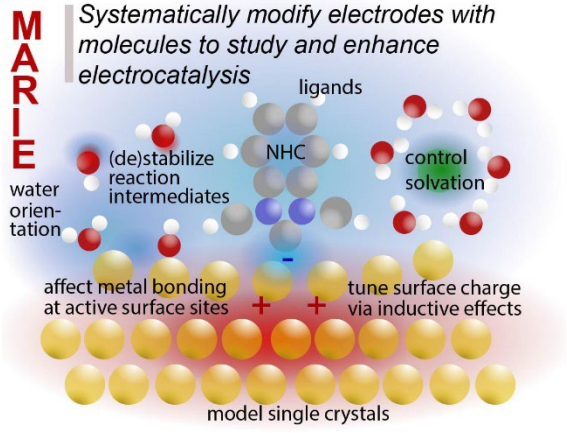
Controlling chemical reactions at the level of the atoms and charge carriers (electrons) that comprise them is a grand challenge that underlies progress across the Department of Energy (DOE) Energy EarthShots. Electrocatalysis offers an opportunity to drive chemical production and industrial processes with renewable energy, and store this electricity in the form of chemical bonds. The efficiency of these processes, however, is limited by the activity and selectivity of the catalysts that drive them. Historical catalyst design has considered distinct approaches: employing extended solids like metals and their oxides, designing discrete molecules with active metal centers, and utilizing nature's biological approach of enzymes. While each approach offers some benefit, for example ease of scale up or increased tuneability, these come with trade-offs that inherently limit catalyst design. This research will combine these strategies to design catalysts that offer precise control over the reactive interface, manipulating processes spanning from adsorbate binding to solvation effects. The mission of MARIE is to design the local reaction environment at an atomically-precise solid surface (active site) by functionalizing it with designer molecules, leveraging the exceptional stability of new classes of molecules (N-heterocyclic carbenes, NHC) synthesized by the team. These molecules will be designed to control how electrons flow to and from the adjacent active site, and will further manipulate aspects of the surrounding environment including solvent effects and the local electric field. Advanced characterization methods, including those at DOE National Laboratories, will illuminate the link between catalyst characteristics and the resultant effects on the reaction environment. This will be coupled with state-of-the art computational methods simulating the reactive interface to build the rigorous chemical and physical understanding necessary for predictive control and design. In doing so, this research will develop fundamental understanding of important chemical processes for a sustainable future. The MARIE foundational basic-science effort underlies advanced electrochemical hydrogen production, electrochemical conversion of carbon dioxide to value-added products, long-duration electrochemical energy storage, and electrifying-metal-production Energy EarthShot goals. Throughout these research efforts, we will significantly broaden participation in electrochemistry at the K-12, undergraduate, and master's level through designated outreach and training efforts that intentionally and effectively engage traditionally underrepresented populations.



This research was selected for funding by the Office of Basic Energy Sciences.

Center for STrain Optimization for Renewable Energy (STORE)

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To meet the requirements of the Long Duration Storage Energy Earthshot, rechargeable batteries for grid leveling and other grid-related uses are needed. The rising cost of lithium (Li), however, limits the practicality of using Li-ion (Li^+) batteries for this application. One well-considered option is to develop sodium-ion (Na^+) batteries, as sodium (Na) is readily sourced in the US and significantly less expensive than Li. For lower cost Na to replace Li in batteries, however, methods are needed to accommodate the larger size of Na^+ and overcome the detrimental effects of strain from Na^+ insertion. The scientific mission of the Center for Strain Optimization for Renewable Energy (STORE) is to develop methods to mitigate and manage structure and volume change upon Na^+ insertion in electrochemical energy storage materials. The development of such designed materials will lead to lower cost electrodes for sodium-ion battery based grid-level energy storage. The three research projects that comprise the STORE program all focus on developing insertions hosts that can work without degradation or unduly slow ion diffusion. The first design approach aims to use lower cost metal oxides with large, rigid channels that can reversibly accommodate large guests like Na^+ with minimal structure change. Initial studies will emphasize the use of tunnel structured oxides of titanium on the anode side, and of iron and manganese on the cathode side. The second approach focuses on layered transition-metal insertion hosts, which can distort more freely upon Na^+ insertion. This effort is directed at understanding the poorly reversible glide transitions that frequently occur upon Na^+ intercalation. The goal here is to develop structures and chemistries that can either suppress glide transitions or make them reversible. Two complementary strategies are planned: (i) design new host chemistries and electrode particle geometries that accommodate elastic deformations to facilitate reversible glide phase transformations; (ii) determine new material compositions that suppress glide transformations altogether. The third and final project takes a different approach to the challenge of large strains. This effort involves materials with amorphous intermediates that plastically deform to accommodate strains, thus mitigating structural degradation through accommodation of large strains, rather than through suppression. The STORE team is largely geographically localized in Southern California, and brings together unique experimental expertise in key areas, including novel materials synthesis (ranging from nanoscale to bulk materials), and multi-scale structural characterization (using *operando* transmission X-ray microscopy, transmission electron microscopy, and a range of X-ray scattering tools). Theoretical efforts, which combine machine-learning approaches with first-principles statistical mechanics simulations, can create predictive frameworks for modeling disordered materials. At the continuum scale, variational formulations, including phase-field models are coupled with understanding gained from first-principles simulations to provide insights into the interplay between local structure, fracture, and ion diffusion.

This research was selected for funding by the Office of Basic Energy Sciences.

Atomic Level Compositional Complexity for Electrocatalysis (Atomic-C^2E)

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Electrocatalytic conversions of CO_2 into value-added chemical fuels and hydrogen production via water electrolysis are essential avenues with enormous potentials to fulfill the goals of zero net carbon emission and reduced environmental impacts, while meeting the energy demand and improving the quality of life. In a research environment where diversity is valued, equity is ingrained, and inclusion is integral, the center of *Atomic Level Compositional Complexity for Electrocatalysis, Atomic -C^2E*, will integrate fundamental electrochemistry, ab initio quantum chemical and multi-scale simulations, and materials chemistry to develop a mechanistic understanding of the CO_2 reduction reaction (CO_2RR) and the limiting step/s during water electrolysis in the O_2 evolution reaction (OER). Acquiring this knowledge will directly support the goals of the Hydrogen and Carbon Negative DOE Energy Earthshots. The elucidated fundamental insights will address technological bottlenecks challenging current electrocatalysts, such as high cost, poor stability, limited efficiency, and/or selectivity of value-added products. Three interconnected thrusts will be pursued with a focus on achieving an atomic-level understanding. The thrusts' aims are to **1)** Elucidate the effects of high compositional heterogeneity in electrocatalytically active surfaces, specifically high entropy oxides for OER in acidic environments. This thrust will investigate how strong compositional heterogeneity can be used to promote robust electrocatalytically active surfaces; **2)** Correlate the compositional and structural rigor and tunability with CO_2RR electrocatalytic activity and selectivity, i.e., the relative kinetics and proximity effects in atomically precise nanoclusters. Copper and gold will be the primary metals of interest to boost the production efficiency and selectivity of value-added products; **3)** Develop and apply transferable multiscale computational models with experimental feedback, and to aid and guide experiments transcending specific material types. *Atomic-C^2E* is led by Georgia State University (GSU), a Predominantly Black and Hispanic Serving Institution in downtown Atlanta. GSU is within about 2.5-hour drive to Oak Ridge National Laboratory (ORNL), enabling effective and efficient outreach to underrepresented workforces and hands-on access to state-of-the-art facilities as elaborated in a Promoting Inclusive and Equitable Research (PIER) Plan.

This research was selected for funding by the Office of Basic Energy Sciences.

Advanced Multi-Physics Machine Learning for Subsurface Energy Systems Across Scales

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Climate change is a pressing crisis, and the Department of Energy's (DOE) Energy Earthshots initiative holds the potential to drive transformative innovations for its mitigation. An interdisciplinary team from five institutions (academic, national lab, and industry) with expertise in diverse disciplines (applied mathematics, data science, geochemistry, geoscience, and engineering) is uniquely positioned to contribute to this initiative significantly. The team will focus on the Enhanced Geothermal Shot and the Carbon Negative Shot.

This project aims to build a computational model for a comprehensive understanding of physical, geomechanical, and geochemical processes occurring at multiple scales in subsurface energy storage and recovery systems. The resulting model will facilitate more accurate predictions and the ability to optimize and manage enhanced geothermal and carbon sequestration systems. To achieve this goal, the team will develop innovative multi-physics machine learning techniques integrated with data assimilation that bridge the gaps among scales. This project will incorporate state-of-the-art laboratory facilities to conduct experiments, the data from which will then be utilized in simulations. To ensure the robustness and reliability of the developed framework, the collaborative team will validate the model predictions and quantify uncertainties in the model by conducting thorough characterizations and analyses of the experimental and field data. The team is committed to promoting diverse, equitable, inclusive, and accessible work environments, aligning seamlessly with DOE values.

This research was selected for funding by the Office of Advanced Scientific Computing Research.
