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Laboratory Directed Research and Development Program

FY 2023 COMPLETED PROJECTS

OAK RIDGE National Laboratory

ORNL/PPA-2024/2

Oak Ridge National Laboratory

LABORATORY DIRECTED RESEARCH AND DEVELOPMENT PROGRAM

FY 2023 COMPLETED PROJECTS REPORT

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CONTENTS

ABBREVIATED TERMS

2.5D	2.5-dimensional
282	Haynes 282
4-HT	4-hydroxy-2,2,6,6-tetramethylpiperidin-1-oxyl, or 4-hydroxy-TEMPO
AFM	atomic force microscopy
AI	artificial intelligence
AL	active learning
ALICE	A Large Ion Collider Experiment
AM	additive manufacturing
APPL	Advanced Plant Phenotyping Laboratory
AQS	anthraquinone-2-sulfonate
bcc	body-centered cubic
BDS	broadband dielectric spectroscopy
CBS	concrete biological shield
CCC	Custom Correlation Coefficient
CCM	carbonated cementitious material
CCM	central carbon metabolism
CCUS-T	Carbon Capture Use Sequestration and Transportation System
CMC	ceramic matrix composite
CMT	cold metal transfer
CNMS	Center for Nanophase Materials Sciences
CNN	convolutional neural network
CREATE	CRISPR-enabled trackable genome engineering
cryoEM	cryogenic electron microscopy
CTE	coefficient of thermal expansion
CV	cyclic voltammogram
DAC	direct air capture
DBU	1,8-diazabicyclo[5.4.0]undec-7-ene
dc	direct current
DCLL	dual-coolant PbLi
DED	directed energy deposition
DISK	Distributed Internet of Things Systems Key Management
DME	dimethyl ether
DFT	density functional theory
E3SM	Energy Exascale Earth System Model
EBC	environmental barrier coating
EBIE	electron beam–induced etch
EIC	Electron Ion Collider
ELM	E3SM Land Model
EMCal	Electro Magnetic Calorimeter
ePIC	Electron-Proton/Ion Collider
eQTL	expression quantitative trait locus
ERL	energy recovery linac
ESA	electrical swing adsorption

EV	electric vehicle
EVI	Enhanced Vegitation Index
fcc	face-centered cubic
FCN	ferrocyanide
FEL	free-electron laser
FFA	fixed-field alternating gradient
FIB	focused ion beam
FOCal	Forward Calorimeter
FPP	fusion pilot plant
FW/B	fusion wall/breeder
FY	fiscal year
GAN	generative adversarial network
GC	gas chromatograph
GCNN	graph convolutional neural network
GOSIF	Global OCO-2 (Orbiting Carbon Observatory-2) based SIF (solar-induced chlorophyll fluorescence)
GPU	graphics processing unit
gRNA	guide RNA
GWAS	genome-wide association study
HCal	hadronic calorimeter
HFIR	High Flux Isotope Reactor
HiDAC	hybrid direct air capture contactor
HN	Hastelloy-N
HPC	high-performance computing
ICP	inductively coupled plasma
ICU	intensive care unit
IEEE	Institute of Electrical and Electronics Engineers
IGBT	insulated gate bipolar transistor
INS	inelastic neutron scattering
INTERSECT	Interconnected Science Ecosystem
IoT	Internet of Things
IR	infrared
iRF-LOOP	iterative random forest leave-one-out-prediction
ISP	isostatic pressing
K-Sar	potassium sarcosinate
LDES	long-duration energy storage
LFHCal	longitudionally segmented forward hadronic calorimeter
LHC	Large Hadron Collider
LIBS	laser-induced breakdown spectroscopy
LiTFSI	Li bis(trifluoromethanesulfonyl)imide
LM	liquid metal
LSCF	La Sr Co ferrite
LSNT	Ni-doped La Sr titanate
LTT	low-temperature transformation
MD	molecular dynamics
MEMS	microelectromechanical
MHDVs	medium- and heavy-duty vehicles
ML	machine learning
MNIST	Modified NIST
MOF	metal–organic framework
MOOSE	Multiphysics Object-Oriented Simulation Environment

MPEX	Material Plasma Exposure eXperiment
MS	mass spectrometry
NCCPI	National Commodity Crop Productivity Index
NDIP	Neutron Data Interpretation Platform
Ni–GDC	Ni supported over Gd-doped ceria
NMC622	$LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$
NMR	nuclear magnetic resonance
NNP	neural network potential
NP	nanoparticle
NPP	net primary production
OCM	oxidative couplilng of methane
OMP	optical potential model
OPSI-MS	open-port sampling interface–mass spectrometry
PB	Prussian blue
PEI	polyethylene imine
PMI	plasma–material interaction
PoL	pattern of life
PSS	polystyrene sulfonate
PVA	polyvinyl alcohol
PVC	polyvinyl chloride
PWM	pulse-width modulation
QCD	quantum chromodynamics
QGP	quark–gluon plasma
QMC	quantum Monte Carlo
QML	quantum machine learning
QUBO	quadratic unconstrained binary optimization
R&D	research and development
R/N	radioactive/nuclear
RAFM	reduced-activation ferritic–martensitic
RCCA	refractory complex concentrated alloy
RCM	reflectance confocal microscopy
RF	radio frequency
RFB	redox flow battery
RL	reinforcement learning
RT	room temperature
SAR	sequence-to-activity relationship
SE	solid electrolyte
SiPM	Si photomultiplier
SKIP	Skel Intelligent Planner
SNP	single nucleotide polymorphism
SNS	Spallation Neutron Source
SOC	soil organic carbon
SOEC	solid oxide electrolysis cell
SP	single-particle
SRF	superconducting radio frequency
SSA	specific surface area
SSB	solid-state battery
SSL	self-supervised segmentation
SSURGO	NRCS Soil Survey Geographic Database
TD-MS	thermal desorption–mass spectrometry
TEM	transmission electron microscopy

TEMPO	tetramethylpiperidine-1-oxyl
TENDL	TALYS Evaluated Nuclear Data Library
Ti–NZSP	Ti-doped $Na_3Zr_2Si_2PO_{12}$
TL	transfer learning
TOF	time of flight
TSLL	toroidally symmetric PbLi
UQ	uncertainty quantification
UTS	ultimate tensile strength
VAE	variational autoencoder
WoSIS	World Soil Information Service
WPT	wireless power transfer
XFEL	x-ray free electron laser
XRD	x-ray diffraction
YDS	$Y_2Si_2O_7$
YS	yield strength

Federal Organizations

DOE	US Department of Energy
EPA	US Environmental Protection Agency
NRCS	US Department of Agriculture Natural Resources Conservation Service
NAS	National Academies of Science
NIST	National Institute of Standards and Technology
TVA	Tennessee Valley Authority

DOE Laboratories and Facilities

CEBAF	Continuous Electron Beam Accelerator Facility
Jefferson Lab	Thomas Jefferson National Accelerator Facility
NETL	National Energy Technology Laboratory
ORNL	Oak Ridge National Laboratory

DOE Offices and Programs

AMMTO	Advanced Materials and Manufacturing Technologies Office
ARPA-E	Advanced Research Projects Agency—Energy
ASCR	Office of Advanced Scientific Computing Research (office and program)
BER	Office of Biological and Environmental Research
BERAC	Biological and Environmental Research Advisory Committee
BES	Office of Basic Energy Sciences
BESAC	Basic Energy Sciences Advisory Committee
BETO	Bioenergy Technologies Office
BRN	Basic Research Needs
BRN-Micro	BRN Microelectronics
BRN-NextGenQS	BRN Next-Generation Quantum Systems
BRN-QMEnergy	BRN Quantum Materials for Energy Relevant Technology
BTO	Building Technologies Office
EERE	Office of Energy Efficiency and Renewable Energy
FEMP	Federal Energy Management Program
FES	Office of Fusion Energy Sciences
FESAC	Fusion Energy Sciences Advisory Committee
HEP	Office of High Energy Physics
HFTO	Hydrogen and Fuel Cell Technologies Office
LDRD	Laboratory Directed Research and Development

MSE	Materials Science and Engineering Division
NE	Office of Nuclear Energy
SC	Office of Science
SPP	Strategic Partnership Projects
VTO	Vehicle Technologies Office
WETO	Wind Energy Technologies Office
WPTO	Water Power Technologies Office

INTRODUCTION

Oak Ridge National Laboratory (ORNL) is the US Department of Energy's (DOE's) largest multiprogram science, technology, and energy laboratory. It possesses distinctive capabilities in a variety of fields, such as neutron science, computing, advanced materials, and nuclear science and technology. Using these capabilities, ORNL conducts basic and applied research and development (R&D) to support DOE's overarching mission "to ensure America's security and prosperity by addressing its energy, environmental and nuclear challenges through transformative science and technology solutions."[1] As a national resource, ORNL also applies its capabilities and skills to the specific needs of other federal agencies and customers through the DOE Strategic Partnership Projects (SPP) Program. Information about the laboratory and its programs is available on the ORNL website.[2]

The Laboratory Directed Research and Development (LDRD) Program at ORNL operates under the authority of the DOE Order 413.2C, *Laboratory Directed Research and Development*,[3] which establishes DOE's requirements for the program while providing the laboratory director broad flexibility for program implementation. The LDRD Program funds are obtained through a charge to all laboratory programs. Although it represents a relatively small portion of the overall research budget, the LDRD Program plays an essential role in maintaining the laboratory's ability to respond to national needs. The program allows ORNL to improve its distinctive capabilities and to enhance its ability to conduct cutting-edge R&D. In accordance with the DOE order, R&D projects funded through the LDRD Program at ORNL support the goals of

- maintaining the scientific and technical vitality of the laboratory;
- enhancing the laboratory's ability to address future DOE missions;
- fostering creativity and stimulating exploration of forefront areas of science and technology;
- serving as a proving ground for new concepts in R&D; and
- supporting high-risk, potentially high-value R&D.

This report provides an overview of the LDRD Program at ORNL in FY 2023 and contains summaries of all the LDRD research projects that concluded between October 1, 2022, and September 30, 2023.

[1] DOE. "Mission." https://www.energy.gov/mission
[2] ORNL. "Home page." https://www.ornl.gov/
[3] DOE. *Laboratory Directed Research and Development*. DOE Order 413.2C. Washington, DC: US Department of Energy. https://www.directives.doe.gov/directives-documents/400-series/0413.2-BOrder-C/@@images/file

LDRD Program Summary

ORNL has established a program with five complementary subprograms to meet its LDRD objectives and to meet the particular needs of the laboratory. A provision for multiple routes of access to ORNL LDRD funds maximizes the likelihood that novel ideas with scientific and technological merit will be recognized and supported.

The LDRD Program at ORNL comprises five subprograms:

- The **Director's R&D Program** supports projects that advance research frontiers, capabilities, and expertise at ORNL in key strategic areas.
- The **Seed Program** supports innovative high-risk/high-reward research to the proof-of-principle stage.
- The **Strategic Hire Program** supports the research of key new staff whose expertise and capabilities address a critical strategic need for the laboratory.
- The **Distinguished Staff Fellowships** assist the laboratory in bringing in exceptional early-career scientists to refresh and expand its scientific and technical expertise.
- The **Early Career Competition** invests in early-career scientists and engineers to cultivate new research leadership.

The total ORNL LDRD Program expenditure of $62.3 million was approximately 3.2% of the laboratory's total budget of $1,929,662,400 for operating and capital expenses, which is well below the maximum of 6% allowed by DOE Order 413.2C and is in accordance with Section 309 of Division D of the Consolidated Appropriations Act.[4]

Table 1. ORNL FY 2023 LDRD allocations

	Director's R&D Program	Seed Program	Strategic Hire Program	Distinguished Staff Fellowships	Early Career Competition	Total
Cost ($)	47,534,015	5,305,827	6,238,628	2,794,375	460,208	62,333,053
Number of projects	98	42	28	15	8	191

Director's R&D Program

The development of research initiatives is the primary tool by which the laboratory develops, builds, and enhances capabilities in particularly promising areas of science and technology to meet anticipated national needs. The Director's R&D Program plays an important role in helping ORNL realize successful outcomes for its initiatives by providing resources to conduct R&D in key areas.

In FY 2023, the Director's R&D Program supported research under the following initiatives, which aligned with the ORNL Lab Plan and emerging sponsor needs:

- Accelerating Radiotherapeutics through Advanced Molecular Constructs
- Artificial Intelligence (AI)
- Emerging and Cyber Security Technologies
- Enzyme Engineering
- Integrated Studies of Complex Biological and Environmental Systems
- Materials Innovation: Digital Metallurgy
- Neutron Data Interpretation Platform Ecosystem

[4] Consolidated Appropriations Act., Public Law 113-76. Washington, DC: US Congress, (2014). https://www.congress.gov/113/plaws/publ76/PLAW-113publ76.htm.

- Nonproliferation Science
- Self-Driven Experiments for Science/Interconnected Science Ecosystem (INTERSECT)
- Transformation Energy Science and Technology
- Transformational Decarbonization
- Validated Design and Evaluation of Fusion Wall Components
- Vertex: Advanced Multiphysics Simulations for Core Applications

Additionally, the program supported a small number of projects outside of the focused initiatives. These projects, selected to address emerging strategic needs, are called Deputy Director Discretionary projects.

The research priorities for each of the initiatives are described in the following sections.

Accelerating Radiotherapeutics through Advanced Molecular Constructs

Recent years have seen a dramatic increase in the medical application of radioisotopes as imaging, diagnostic, and therapeutic agents. This increase is driven partly by the increased use and acceptance of biologicals as therapeutics for cancer, infectious diseases, and inflammatory disorders. Current excitement around targeted radiotherapy combines advances in the development of biological agents with the ability to arm these disease-targeting biomolecules with a nuclear payload (i.e., α-, β-, or Auger-emitting radioisotopes). The convergence of radioisotope production with the rise of biologicals to treat disease requires advances in chemical and nanomaterial technologies to join the two and achieve the most effective radiotherapeutics. This precision medicine approach will revolutionize medicine and will lead to targeted combination therapies. This initiative, drawing on ORNL's strong R&D program in radioisotope production for medical applications, engages multiple scientific disciplines to develop fundamental understanding of the inherent chemistry and biological effects of radiotherapeutics at the subcellular to multicellular and tissue scales.

This initiative focuses on research in three areas.

- **Ultrachelators:** understanding the coordination chemistry of actinide, lanthanide, and alkaline earth radioisotopes to guide in silico design and synthesis of chelators for optimal radiometal binding and stability
- **Nanoconstructs:** developing nanoconstructs for theranostics and combination therapeutics (radiotherapy, chemotherapy, and immunotherapy) and to leverage precision medicine approaches for cancer imaging, diagnostics, and treatment
- **Targeting vectors and radiobiology:** improving precision targeting of radionuclides to cancer cells and understanding of the mechanism of action of therapeutic radionuclides at the target site.

Artificial Intelligence

The founding goals of the AI Initiative were to conduct fundamental R&D in AI to enable next-generation science and engineering. In FY 2023, the initiative focused on the development of computationally efficient surrogates for multiscale systems and processes, the development of AI-based methods for the design and optimization of these systems, and the development of machine learning (ML) models to represent complex engineered and physical systems, as well as the use of those models for performance prediction, prognostics, and control of these systems. Key considerations included robustness (in learning, control, design, and decisions), dimension reduction (in creating surrogates or digital twins), and computational efficiency (in the use of high-performance computing [HPC] or edge resources).

Emerging and Cyber Security Technologies

This initiative focuses on developing foundational capabilities that enable effective cyber defense as new and emerging technologies create opportunities to enhance cybersecurity and new potential vulnerabilities. Topics of interest include

- developing appropriate scalable and multimodal cyber architectures capable of processing and analyzing cyber events from the edge, Internet of Things, or fog;
- developing robust and adaptable cyber defense protocols that ensure the fidelity of hybrid quantum–classical systems for sensing, communication, and computation;
- applying, customizing, and developing appropriate AI-based cyber techniques capable of dynamic adaptation and autonomic response to novel cyber threats; and
- creating resilience to adversarial exploitations of AI, which are effective means of circumventing AI-based cyber defenses.

Enzyme Engineering

The Enzyme Engineering Initiative seeks to rapidly engineer enzymes that catalyze reactions at material interfaces. Enzymes can perform challenging chemical transformations with high specificity under benign conditions. Natural enzymes can be further improved through directed evolution to improve activity, specificity, and stability. However, current enzyme engineering methods are slow and, largely because of analytical limitations, are difficult to use with enzymes that act at material interfaces. Addressing these limitations will enable novel biochemical solutions to challenges in materials science. This initiative pursues computational enzyme design using ML, high-throughput enzyme expression, rapid analytical techniques, and atomistic simulations focused on polymer hydrolysis and protein binding of Pt group elements.

Integrated Studies of Complex Biological and Environmental Systems

A large proportion of microbial and plant genes have unknown functions, and characterized genes often cannot be linked to functional traits. These knowledge gaps limit the ability to develop mechanistic models that accurately simulate an organism's activities and response to change in environmental conditions. Furthermore, although traits have become fundamental to understanding ecology and evolution, predicting how these traits affect large-scale systems, such as ecosystem, watershed, and Earth-system levels, remains a grand challenge. Global change may elicit unusually large feedback from some of these ecosystems, for which vulnerability and response are uncertain. Addressing this challenge will accelerate research for critical needs in bioenergy and bioproducts, biosystems design, carbon capture technologies, and the predictability of ecological and Earth systems. This initiative focuses on advancing the understanding, design, and engineering of the fundamental processes influencing complex systems' response, resilience, and recovery to extreme environmental and climatic events. To accomplish this objective, the initiative explicitly targets multiple scales: cells, organisms, communities, and ecosystems. The focus on resiliency acknowledges that biological and ecological systems possess an incredible potential to adjust and adapt to wide fluctuations in their surrounding environments.

This initiative supports the following key areas of research:

- Mechanistic insights into the interfaces among microbiology, geochemistry, and root–soil processes in the coupling of nutrient and trace metal cycling and how biotic–abiotic interactions translate from the molecular level to genetic and ecosystem scales
- High-throughput genomic characterization, functional validation, and phenotyping that supports work at the intersection of trophic scales and kingdoms
- Coupling terrestrial and aquatic systems to numerically describe how climate, land use and cover, and trace element fate and transformation influence stream biogeochemistry
- Modeling studies to identify threshold to ecosystem resiliency (e.g., when ecosystems reach a tipping point to thermal perturbations) and to determine what aspects of ecosystems are most and least resilient to extreme perturbation

Materials Innovation: Digital Metallurgy

The Materials Innovation initiative focuses on conceptualizing materials solutions for applications in extreme environments, addressing the multifaceted challenges necessary to bridge the gap between metal-alloy design and additive approaches to processing and product manufacturing. This approach, termed *digital metallurgy*, combines cutting-edge concepts in physical metallurgy, computational alloy design, advanced manufacturing, and software control to arrive at unprecedented materials solutions. The emphasis is on the development of extreme materials that cannot be produced with current approaches and the development of novel approaches to building structural materials that short-circuit the traditional approach of sequential materials and product development.

Topics of interest include the following:

- Fundamental studies that leverage and demonstrate a digital basis for extreme environment-resistant microstructure design through advanced manufacturing of materials
- Studies that combine the geometrical freedom associated with additive manufacturing with the metallurgical complexity that is harnessed by physical metallurgists with an understanding of microstructure–property relationships
- Microstructure design research with an emphasis on phase transformations, grain structure, digital functional grading, and other metallurgical phenomena for improvement of specific properties of relevance for applications in extreme environments
- Materials design strategies that overcome traditional trade-offs in properties such as strength and ductility, with materials of interest including high-entropy alloys and refractory alloys

Neutron Data Interpretation Platform Ecosystem

The High Flux Isotope Reactor and Spallation Neutron Source provide world-leading experimental capabilities to a diverse scientific user community. Interpretation of neutron data is necessary to convert measurements into scientific knowledge. As problems under investigation increase in complexity and neutron instrumentation provides larger data volumes with more sophisticated experiments, simple approaches to data analysis are often insufficient, and advanced modeling and analysis methods are required to generate the most effective science from these neutron sources. The purpose of this initiative is to push the boundary of advanced neutron data analysis and modeling and lay the foundation for future developments including autonomous or guided experiments. Projects in this initiative focus on three areas.

- **Real-time data analysis:** Develop tools and workflows to enable automated data analysis. This analysis will enable scientific content to be extracted rapidly from neutron data and will lay the foundation for automated or guided experiments.
- **HPC materials modeling:** Incorporate computationally intensive materials modeling approaches into data interpretation workflows. Modular workflow implementation will enable the development of digital twins of neutron instruments, which will allow for virtual experiments to be performed and synthetic data to be generated.
- **AI/ML applications:** ML approaches have shown potential for advancing the understanding of complex datasets, such as those generated at the Spallation Neutron Source. These methods can enable feature identification, background and artifact suppression, materials and signal classification, property prediction, and other activities. New approaches and workflows will be developed to enable the rapid, automated interpretation of neutron scattering data with the potential for experiment optimization or guiding.

Nonproliferation Science

The Nonproliferation Science Initiative advances fundamental scientific understanding and develops applications needed for the security of the nation. This initiative is structured around two crosscutting,

foundational thrust areas in nuclear nonproliferation research: (1) pattern of life (PoL) and (2) signal detection in low signal-to-noise data. The PoL thrust is focused on demonstrations of significant improvements to existing PoL analysis techniques and of new types of PoL analysis on facilities gleaned using emerging sensing techniques or algorithms, as well as new visualization and validation techniques for PoL analysis. The signal detection thrust is focused on the ability to detect and/or verify operational processes related to the nuclear fuel cycle. Research under this thrust focuses on advanced, ab initio, computational models of large, complex, solid-state actinide systems; novel methods for extracting chemical/physical information connecting material properties to formation; and novel data analyses designed to search for new signatures.

Self-Driven Experiments for Science/Interconnected Science Ecosystem

The objective of this initiative is to develop the capabilities needed to automate entire scientific workflows and control them with AI/ML to bring about revolutionary efficiencies, allowing researchers to explore high-dimensional problems previously considered impossible and discover new, subtle correlations. INTERSECT focuses on two key areas of development: automation (incorporates tools that perform well-defined, systematic processes with limited human intervention) and autonomy (smart decision-making techniques, such as AI and ML) of processes, experiments, and laboratories, including the following:

- Creating new, automated methods and tools that incorporate on-the-fly data analysis, providing autonomous and rapid feedback that steers the course of a synthesis workflow
- Integrating automation with experimental synthesis techniques
- Combining ORNL biological and Earth science capabilities with high-throughput edge computing to facilitate data collection, interpretation, and autonomous labs
- Integrating advanced manufacturing instruments, sensors, and computational resources (e.g., edge nodes and/or HPC)
- Developing multidomain interconnected laboratory capabilities, including building equipment, renewable energy generation, energy storage, and electric grid, to accelerate large-scale integration of clean energy for utility-scale decarbonization and energy resilience
- Integrating AI-based approaches to the design and steering of experiments for scientific discovery or for the design of new systems, parts, materials, or processes
- Integrating reinforcement learning techniques to demonstrate autonomous control of instruments, facilities, or smart laboratories

Transformation Energy Science and Technology

The Transformation Energy Science and Technology Initiative works on the frontier of the science of novel battery concepts for the electrification of transportation and the grid through synergistic breakthroughs in materials R&D, battery engineering, modeling, and advanced operando characterization. The research focus areas for this initiative include the following.

- **Solid-state batteries (SSBs) for the electrification of transportation:** All SSBs, boasting high energy densities and enhanced safety standards, are poised to be the next major advancement in energy storage technologies. Despite that promise, practical deployment of such battery systems remains a distant reality owing to several material- and device-level challenges at each interface in a typical cell configuration. Engineering challenges added to the inherent cell-level challenges pose significant bottlenecks that require a thorough, systematic assessment for effective mitigation to facilitate the practical deployment of SSBs. The solid electrolyte is often considered the linchpin to these batteries; thus, research within this initiative addresses various challenges plaguing present-day solid electrolyte systems at the material- and electrode-processing and device levels.
- **Long-duration batteries for the electrification of grid:** The existing energy storage technologies cannot meet cost, duration, and location requirements of renewable energies. Multipronged research

is needed to develop and demonstrate novel energy storage systems that have the potential to enable the massive deployment of renewable energy technologies. This initiative pursues the development of battery systems that use Earth-abundant resources and all-time rejuvenated reactive components with the potential to provide deep cuts in CO_2 emissions.

Transformational Decarbonization

The Transformative Decarbonization Initiative's objective is to develop innovative concepts and processes for CO_2 capture and/or conversion and electrochemistry for displacing chemical processing traditionally reliant on fossil fuel–derived feedstocks and heat, with an emphasis on three key areas:

- Developing energy-efficient and scalable processes for CO_2 capture from dilute sources (including atmospheric air, ocean water, and flue gas from natural gas turbines) and for a variety of end uses (including geologic sequestration and conversion to value-added products)
- Pursuing materials and process innovation in electrochemistry that can enable or facilitate CO_2 capture and utilization in the context of large-scale chemical and fuel production processes, including research on nonaqueous electrolytes and other nontraditional reaction media, the development of novel chemical cycles, and exploration of device fabrication methods that create avenues for rapidly increasing the scale of operation and for batch/dynamic operations suited to intermittent energy sources
- Defining data-driven approaches to enhance stability/durability of materials for CO_2 capture and utilization to improve the economic viability of such processes with respect to the degradation of materials and device performance over time, with studies focusing on making exemplar materials more robust or on introducing means to regenerate materials

Validated Design and Evaluation of Fusion Wall Components

This initiative combines fusion and fission expertise to accelerate the deployment of fusion energy as part of the US mid- to long-term energy portfolio. The initiative focuses on integrated fusion component modeling that addresses the question of how scientists can predict and control the phenomena around a fusion wall component's plasma–material interface. Work in this initiative is directed toward filling a well-known gap and need for well-verified, well-validated modeling and simulation with the inclusion of uncertainty quantification for fusion components. Its outcomes are intended to be the foundation for the eventual deployment of a pilot plant and to facilitate the intermediate step of validating the critical technology for the pilot plant. The ultimate goal of this initiative is to develop comprehensive models and sensor technology integrated into efficient workflows, allowing validation testing at facilities such as the Material Plasma eXperiment, a fusion prototypic neutron source and a component high–heat flux facility.

Projects in this initiative fit within four thrust areas focused on answering the fundamental science question of how to predict and control the phenomena around a fusion wall component's plasma–material interface, including the following.

- **Thermal management at extremes:** Investigate and develop new methods for thermal management that incorporate real-time feedback control of a fusion component.
- **Plasma–material interactions:** Develop a further understanding of material erosion, migration, and plasma–material interactions.
- **Fusion neutron effects and damage:** Build a foundational basis for blanket technology and new plasma-facing and structural material that can sustain 14 MeV neutrons.
- **Computation:** Introduce computational approaches for integrated performance and response of the plasma–material interface that simultaneously incorporate all three of these science areas.

Vertex: Advanced Multiphysics Simulations for Core Applications

This initiative aims to establish a new computational and data science–ready framework that simulates coupled multiscale/multiphysics phenomena in core mission spaces. An advanced predictive capability is being created in which new individual and coupled physics are developed to meet science-driven needs and demonstrated on problems residing in the areas of (1) next-generation enrichment science for applications, including the plasma separation process, gas centrifuge, and electromagnetic isotope separation; (2) fusion science, including blanket design and the evaluation of novel fusion concepts; and (3) advanced scientific computing workflows integrating data-based models.

Seed Program

The Seed Program supports innovative ideas that have the potential to enhance the laboratory's core scientific and technical capabilities. It also provides a path for funding new approaches that fall within the distinctive capabilities of ORNL but outside the more focused research priorities of the major laboratory initiatives.

Strategic Hire Program

The Strategic Hire Program was formed to add critical skills to the laboratory by hiring individuals whose research is aligned with ORNL's strategic needs. Candidates for strategic hires are expected to be established investigators who are well-qualified for leading research programs, capable of developing substantial programs, and/or able to take organizational leadership roles. Promising individuals at earlier stages of their careers who bring expertise critical to major ORNL strategic initiatives may also be supported.

Distinguished Staff Fellowships

The fellowship programs were formed to provide research opportunities for exceptional early-career scientists in honor of Dr. Eugene Wigner, the first director of R&D at ORNL; Dr. Alvin Weinberg, a former ORNL director; and Dr. Liane Russell, ORNL's groundbreaking geneticist. The appointment of fellows at ORNL provides an opportunity for outstanding scientists and engineers in life, physical, computer, computational, and social sciences to pursue research in areas related to national energy problems and interests. Awardees of Distinguished Staff Fellowships receive funding for their research projects through the LDRD Program.

Early Career Competition

Introduced in FY 2023, the LDRD Early Career Competition cultivates the research and professional development of high-potential, early-career research staff. The competition is open annually with an awardee selected from each of ORNL's eight research directorates.

Report Organization

This report, which provides a summary of all projects that concluded in FY 2023, is divided by subprogram into five sections:

- Director's R&D
- Seed
- Strategic Hire
- Distinguished Staff Fellowships
- Early Career Competition

The Director's R&D Program projects are categorized according to the program initiatives. The Seed Program, Strategic Hire projects, and Early Career Competition projects are categorized by the research

division of the principal investigator. The Distinguished Staff Fellowships are grouped by fellowship. The summaries are arranged within each section by project number; each completed project summary contains

- the project description,
- a brief description of the project's relevance to DOE and ORNL missions, and
- results and accomplishments.

SUMMARIES OF PROJECTS SUPPORTED BY THE DIRECTOR'S R&D PROGRAM

ACCELERATING RADIOTHERAPEUTICS THROUGH ADVANCED MOLECULAR CONSTRUCTS

This initiative did not have any completed projects in FY 2023.

ARTIFICIAL INTELLIGENCE

10584: Uncertainty Quantification for Artificial Intelligence Models to Automatic Neutron Scattering Data Screening

D. Lu, H. Tran, S. Liu, D. Womble, J. Yin, M. Stoyanov, P. Zhang, C. Hembree

Project Description

This project developed an accurate, scalable, and reliable uncertainty quantification (UQ) algorithm for machine learning (ML) models. Existing UQ methods have some limitations in scientific ML. Most of the methods quantify uncertainty on ML model parameters and can incorporate physics to only a limited degree, they usually involve data distributional assumptions and cannot be generally applied in a variety of scientific domains, and they are typically computationally expensive and hard to scale. The team developed a prediction interval method, called PI3NN, to quantify model prediction uncertainty by training three neural networks. The PI3NN method provides understandable information for decision-making by directly communicating uncertainty within lower and upper bounds. It is particularly useful for scientific ML because it is computationally efficient, does not involve distributional assumptions on data, has stable training, and produces reliable performance. The team applied PI3NN for material–property prediction based on the ML models developed in the AI for Scientific Discovery thrust of the AI Initiative. Results indicated that PI3NN can precisely quantify the uncertainty of material–property prediction and that it can identify out-of-distribution samples to guide data collection.

Mission Relevance

This research forms the foundation of UQ for ML models, which can be used for accurate and credible predictions in natural sciences and safe operation and control in complex engineered systems. The team demonstrated its applications in water resources management under the changing climate, safe geological carbon storage decision-making, anomaly detection and prediction in smart grid systems, and material–property prediction and design. These applications align with DOE and ORNL missions and cover different DOE offices and programs, including SC, FECM, and EERE.

Results and Accomplishments

This project developed a novel UQ method, PI3NN, for quantifying ML model prediction uncertainties for trustworthiness. The team demonstrated the developed UQ algorithm in a variety of scientific applications, including accurate material–property predictions, and guided the sample collection to further improve the model prediction accuracy; improved streamflow prediction in the changing climate to inform the reservoir operators' decision-making; accurately estimated geological properties for safe carbon storage management; and accurately predicted transient source locations in a smart grid system.

10676: Towards Artificial Intelligence Augmented Simulations for Accelerated Discovery

G. Cong

Project Description

As artificial intelligence (AI) methods become increasingly adopted as key components of scientific simulations, this project aimed to create a framework to be deployed on modern platforms where online learning steers, guards, and accelerates simulations for better science. In short, the framework supports intelligent simulation, where intelligence includes (1) learning that is tailored for scientific problems and (2) efficient management of the workflows on emerging platforms aided by learning. Toward the ultimate intelligent simulation framework, this project solved two fundamental problems, described as follows.

The first need was a deep learning methodology for scientific domains with rich relationships among entities. Current AI methods in science heavily borrow from text/speech/vision applications. Although effective to a certain degree, these methods do not handle complex relationships and high dimensionality in data generated by simulations very well. This project developed a methodology that leverages such relationships and information for learning purposes. This work combined graph algorithms and graphical neural networks to effectively use the topology information in the graphs for the overall predictive performance.

The second need was for fast, scalable, adaptive, and distributed training with simulation data. Fast training refers to not only time but also convergence and scalability. Large-scale simulations impose requirements such as low communication frequency and fully adaptive solvers, which are absent in current training methods.

Mission Relevance

This project addresses methodologies of leveraging high-performance computing and AI techniques to advance computing in the scientific domains. It directly addresses advanced computing methods as well as applications in materials research.

Results and Accomplishments

This project developed a minimal feature crystallography graph neural network (GNN) that leverages sophisticated architectures rather than domain-specific features for learning. This work showed that this method achieves state-of-the-art performance in comparison with the models with heavily engineered features for a variety of problems, including quantum chemical property prediction of materials and CO_2 adsorption predictions for metal–organic frameworks.

This project introduced a methodology to incorporate classical graph algorithms into learning using GNNs. Using standard benchmarks such as citation networks, this project showed that this methodology significantly improves the performance of transductive learning.

This work investigated generative pretraining for materials with GNNs and compared the pros and cons of several methodologies.

The findings of this project have been published in various journals and conference proceedings.

10792: Atomistic Modeling and Machine Learning for Neutrons

Y. Cheng, Y. Zhang, J. Paddison, G. Sala, M. Stone, A. Savici, D. Pajerowski, A. J. Ramirez-Cuesta, D. Abernathy

Project Description

Data analysis and interpretation is a major bottleneck for the efficiency and productivity at high-flux neutron scattering facilities. High-throughput and high-resolution beamlines combined with increasingly complex samples produce large volumes of complex data that require novel approaches for fast, quantitative, and comprehensive analysis. Moreover, for some new instruments at the Second Target Station, the information-rich output of the broadband measurements necessitates the development of new data analysis approaches. The desired structural and dynamical information is encoded in (multidimensional) neutron scattering data, and a direct forward solution is often difficult. To this end, an atomistic model is an ideal solution to explain the underlying origin of the neutron scattering features. Unfortunately, such models are sometimes challenging to build and analyze. This project addressed these challenges using advanced modeling methods empowered by machine learning analytics. Software and workflows were developed to allow instrument staff and users to perform fast, model-informed data analysis, overcoming existing barriers. An integrated approach to performing calculations parallel to neutron scattering experiments was established, taking advantage of the high-performance computing and edge computing resources available at ORNL. The implementation of this project is expected to build a

strong connection between neutron scattering and computing user facilities at ORNL and significantly boost their scientific output.

Mission Relevance

ORNL hosts two of the most advanced neutron sources in the world, with the third one (the Second Target Station) under development. ORNL also has some of the world's most powerful computing resources, such as Summit and edge computing. This premier hardware must be equipped with world-leading software to achieve its full potential. Combining machine learning and atomistic modeling can be a game changer for many of the issues currently faced in neutron data analysis, and the greatly expanded capability owing to this integration will enable researchers to extract maximum useful information from the neutron data. It will also facilitate users with no modeling background and no special computing resources to perform model-informed data analysis, thus benefiting a broad user community.

Results and Accomplishments

This project has led to the creation of the first large-scale synthetic neutron scattering spectra database. A hybrid neural network, consisting of a symmetry-aware representation of the crystal structure and an autoencoder for spectral data compression, was trained to achieve direct prediction and reconstruction of the neutron scattering spectra from the crystal structure. This data-driven approach significantly accelerated the pipeline from experimental planning and steering to data analysis. A graphical user interface implementing the new capability has been developed and will be deployed.[1] In addition to performing direct predictions, this project demonstrated that a properly trained machine learning force field can achieve excellent accuracy for neutron spectroscopic analysis with more flexibility and transferability. A workflow to develop machine learning force fields for neutron data analysis has been implemented in the Neutrons Data Interpretation Platform[2] and is available to the user community.

10889: Artificial Intelligence-Enabled Association of Plant Physiology and Phenotypes

J. Lagergren, J. Streich, M. Pavicic Venegas, H. Chhetri, M. Cashman McDevitt, V. Melesse Vergara, L. York, D. Jacobson

Project Description

The increased frequency of extreme climatic events is putting unprecedented pressure on biological and ecological systems. These systems have the potential to adapt to such pressures, but understanding the mechanistic processes underlying plant response and recovery is complex. Manually acquiring data on plant traits relevant to plant response and recovery is time-intensive, destructive for many measures, and often requires subject matter expertise and training. A need exists for automated image processing and predictive modeling to infer such traits, or close analogs, directly from available image data to eliminate such bottlenecks, thereby facilitating downstream scientific analysis of plant response and resilience to extreme climatic events. This project proposed the development of a suite of image and data analysis tools to infer biologically relevant plant traits from image data and to draw associations between traits using explainable artificial intelligence and network analysis. The project was divided into three main aims: (1) develop deep learning models for segmentation and feature extraction from images, (2) use explainable artificial intelligence to associate functional modules of plant traits relevant to abiotic stresses, and (3) scale computational methods to accommodate future high-throughput phenotyping systems.

[1] analysis.sns.gov
[2] calvera.ornl.gov

Mission Relevance

To accelerate the development and deployment of solutions to the nation's energy and environmental challenges, the need is increasing to develop sustainable perennial crops that thrive in suboptimal environments and are resilient to abiotic stresses with multiple mission-relevant use cases (e.g., conversion of biomass to advanced bioproducts and sustainable aviation fuel). These goals are facilitated by connecting plant gene functions to observable traits through the automated, high-throughput measurement of key plant characteristics but are bottlenecked by the availability of large, high-quality datasets, computing systems for large-scale image and data analysis, and the development of computational methods that leverage such datasets and computing resources.

Results and Accomplishments

The first aim was achieved by developing convolutional neural networks (CNNs) for semantic segmentation of leaf and vein architecture to enable the extraction of more than 100 leaf traits in *P. trichocarpa*,[3] a mission-relevant bioenergy crop. The CNNs were developed for a dataset of RGB leaf scans that were collected by the team in late FY 2021. In contrast to alternative methods, the developed approaches break down the complex problem of whole-image segmentation into smaller, easier decision rules and require very few samples for training (e.g., just eight images for vein segmentation). The segmentations were used to extract more than 100 leaf and vein traits that were validated with real-world physical measurements and used for downstream genomic analysis. The CNNs were later applied to pseudo-RGB images constructed out of the hyperspectral image dataset, in which segmentations were used to generate a pixel-level dataset composed of spectral distributions. These distributions were used in a regression framework with hyperspectral transformers and gradient-boosting decision trees to predict the corresponding leaf-level physiological measurements. The second aim was achieved by using iterative random forest leave-one-out-prediction (iRF-LOOP) to generate predictive phenotype networks from the CNN-derived leaf traits and extended datasets at the genotype-level, which integrated greenhouse and field data of plants undergoing a range of experimental conditions. The third aim was achieved by converting the CNN segmentation methods for inference to command-line scripts that leverage graphics processing unit–accelerated computing and were demonstrated using an NVIDIA DGX System A100 to segment and phenotype the RGB dataset (totaling 2,906 images) multiple orders of magnitude faster compared with manual annotation. The iRF-LOOP method used the Summit high-performance computing system to construct feature association networks.

This project solved challenges in biological image analysis using artificial intelligence and ORNL computational resources that will increase the ability of researchers to monitor plant physiology and design bioenergy feedstocks.

11068: Active Learning of Atomistic Surrogate Models for Rare Events

G. S. Jung, S. Lee, J. Y. Choi, S. Irle, G. Cong, F. Liu

Project Description

Atomistic modeling with artificial intelligence is an emerging tool for understanding materials' properties and behaviors and predicting novel materials with optimized or targeted properties. Atomistic surrogate models are outstanding in this field because they have shown a comparable accuracy to ab initio electronic structure calculations for reproducing potential energy surfaces while being several orders of magnitude faster. However, such surrogate models can perform poorly outside their training domain and typically fail in predicting rare events in molecular dynamics (MD) simulations. Enhanced sampling and dynamics loading can capture rare events in MD simulations by overcoming an energy barrier. The team developed an automated active learning (AL) capability by combining surrogate models and enhanced

[3] J. Lagergren et al. "Few-Shot Learning Enables Population-Scale Analysis of Leaf Traits in *Populus trichocarpa*." *Plant Phenomics* 5, 2023. DOI: 10.34133/plantphenomics.0072

sampling methods for capturing rare events to derive atomistic surrogate models for targeted applications. The developed AL capability was also used for the distillation of many configurational data sampled from empirical forcefield (low-fidelity model) with enhanced sampling techniques. The distilled data can accelerate training with better performance compared with using all data from the sampled configurations.

Mission Relevance

In many cases, key properties related to dynamic processes and rare events, such as energy barriers for chemical bond breaking and formation, transition temperatures for phase transition, and materials strength, are critical to deriving design principles for new materials. Controlling such rare events and related properties plays a central role in material synthesis and manufacturing processes. For example, the transformative processes in nanophase materials such as atom replacements, defect formation, and heteroatom doping by ion beam irradiation or the plasma synthesis processes with chemical precursors play central roles in the development of novel quantum materials for quantum information science and next-generation carbon-neutral hydrogen storage for clean energy technologies.

Results and Accomplishments

The team developed an automated AL capability, including the generating initial data, sampling new confirmation through neural network potential (NNP) and enhanced sampling, selecting configuration based on uncertainty quantification, augmenting data, and retraining with updated data. The key to the capability is combining NNPs and one of the enhanced sampling methods, steered MD, for capturing bond-breaking events of alkane chains to derive NNPs for targeted applications. The team developed a decision engine based on configurational similarity and uncertainty quantification, used data augmentation for effective AL loops to distinguish the informative data from enhanced sampled configurations, and showed that the generated dataset achieved an activation energy error of less than 1 kcal/mol. Furthermore, the team devised a strategy to alleviate training uncertainty within AL iterations via a carefully constructed data selection process that leverages an ensemble approach.

The developed AL process can be used for a more generalized configuration dataset (as a foundational dataset) for metallic systems. Generative models can swiftly propose promising materials for targeted applications. However, the predicted properties of materials via the generative models often do not match with calculated properties via ab initio calculations. NNPs can expedite the process by providing relaxed structures from the initially generated ones. Nevertheless, acquiring data to train NNPs for this purpose can be extremely challenging because the data must encompass previously unknown structures. The team used extended ensemble MD to secure a broad range of liquid- and solid-phase configurations in Ni, one of the metallic systems. Then, the developed AL process was reduced without significant loss of accuracy. The NNP trained from the distilled data could predict different energy-minimized, closed-pack crystal structures even though those structures were not explicitly part of the initial data. Furthermore, the team demonstrated the transferability of the workflow, and the data could be translated to other metallic systems (Al and Nb) without repeating the sampling and distillation processes.

11086: Efficient Reinforcement Learning Using Transfer Learning

H. Zandi, K. Kurte, K. Amasyali, Y. Liu, J. Munk

Project Description

Model-free reinforcement learning (RL) is a powerful tool that has shown promising results. However, because model-free RL algorithms learn optimal control policies by continuously interacting with their environments, these algorithms require a large amount of data to learn, which limits their applicability in many domains. Many approaches, including transfer learning and data augmentation, have already been proposed to address this challenge in imaging applications. However, a significant need still exists for improving the training and data efficiency of model-free RL for nonimaging, real-world applications. Toward addressing this research gap, this project developed and used a framework-agnostic transfer

learning (TL) approach to improve the efficiency of the RL algorithm by reducing data need and reducing training time. TL was used both in the data augmentation and the RL agent training phase. This method allows the controller to be (1) data-efficient, requiring less observational data and shorter exploration periods, and (2) learning-efficient, improving its decisions for unseen data and environment using the knowledge transfer. In this approach, the RL agent uses the knowledge gained by an external expert in the source domain for other tasks in the target domain with no assumption about a particular domain. This work will allow RL agents to learn faster with less data and fewer interactions with the environment.

Mission Relevance

This approach improved the learning efficiency, robustness, and reliability of RL algorithms. This project also demonstrated the usability of the proposed approach for two different applications in simulation, which were also applied to the control of actual devices and actuators in facilities and smart laboratories. This project enables the application of model-free, RL-based optimization methods for controlling instruments and actuators in the field for various inherently complex, nonstationary applications, such as smart buildings, the smart grid, transportation, robotics, and materials, with a reduced amount of training data. This approach is also aligned with the objective of improving efficiency, including both power and data efficiency, in machine learning.

Results and Accomplishments

This project consisted of four experiments using transfer learning. The team implemented and evaluated TL (1) from a set of water heaters to another water heater, (2) from a set of buildings to another building, (3) from water heaters to a building, and (4) from buildings to a water heater. The result of the TL from a set of buildings to another building was accepted to the 2024 IEEE Power and Energy Society Innovative Smart Grid Technologies North America conference.[4] In the paper, the team reported that the distilled deployment is approximately 16% better than the online and approximately 24% better than the baseline deployments. Also, the distilled deployment was able to cross the baseline right after the deployment, whereas the online deployment could only cross the baseline after approximately the 21st day. For the water heater-to-building TL, the result showed that the distilled approach achieved significantly better than the online approach and performed close to the offline and baseline approaches.

11100: Physics-Informed Three-Dimensional Self-Supervised Zero-Shot Semantic Segmentation with Generative-Adversarial Networks

A. Ziabari, M. G. Meena, A. Tabassum, A. Cheniour, M. Alnaggar, D. Rose, S. V. Venkatakrishnan

Project Description

Because of the lack of high-quality annotated data, as well as the variability of imaging modalities, imaging acquisition parameters, and settings, supervised deep learning approaches are limited in addressing the need for image segmentation in scientific imaging applications. This project proposed to develop a physics-informed, self-supervised, zero-shot deep learning method for 3D semantic segmentation of scientific imaging volumes. The key contributions of this project were (1) a constrained generative adversarial network (GAN) architecture for robust realistic data/label pair generation from physics-based simulations and (2) a self-supervised, zero-shot learning exploiting physics-based GAN generated support images/labels to perform 3D semantic segmentation in scientific imaging volumes.

Mission Relevance

With the abundance of scientific imaging data being generated at ORNL user facilities, the development and usage of advanced image analysis methods that can distill the necessary information from data are

[4] K. Amasyali, Y. Liu, and H. Zandi. "A Transfer Learning Strategy for Improving the Data Efficiency of Deep Reinforcement Learning Control in Smart Buildings." 2024 IEEE Power and Energy Society Innovative Smart Grid Technologies North America, Washington, DC, February 19–22, 2024 (upcoming as of the writing of this report).

highly relevant to the users of those facilities. Artificial intelligence–based approaches are at the forefront of these methods and have shown significant promise for different applications. This work paves the way to developing self-supervised, artificial intelligence–based algorithms to advance the processing and characterization capabilities across a range of scientific applications. It aligns with DOE ASCR's basic research in scientific machine learning mission to "transform science and energy research by harnessing DOE investments in massive data from scientific user-facilities."[5] Furthermore, this work provides a proof of concept for the generation of 3D concrete microstructures through self-assisted learning to support modeling efforts. Representations of the concrete specimen in 3D will eventually help to improve the safety assessment of the concrete biological shield of light-water reactors for a potential nuclear power plant lifetime extension.

Results and Accomplishments

This project first developed and trained a 2.5-dimensional (2.5D) U-Net network with a limited number of annotated data to segment microstructure in real x-ray computed tomography data of irradiated concretes. The segmented volume served as a digital twin in a final element simulation for modeling the crack initiation in concrete under irradiation.[6] The focus was then turned on the development of a constrained GAN to generate realistic-looking, real data that can be used for training the self-supervised segmentation (SSL) network. To that end, this project developed a microstructurally constrained contrastive unpaired GAN (based on CUT-GAN[7]). This network leverages physics-based, simulated data as a constraint and an ensemble learning approach to create realistic-looking microstructures that are more faithful to the true structure of the input-simulated data while style-transferring the noise and realistic texture of artifacts into the data.

The trained, constrained GAN was used to generate 800 volumes 256 × 256 × 256 for training the SSL network that was adopted from Ouyang et al. (SSL-AlpNet).[8] The team first tuned the hyperparameter of the SSL-AlpNet network and trained the network on a real dataset without labels (as described in Ouyang et al.). The team called this network a *superpixel algorithm* because it was trained based on labels extracted by the superpixel algorithm. Then, researchers used simulated data and, finally, GAN data to train the same network architecture. The trained network was then used to test on 35 out-of-distribution real data. During the inference, one dataset (a volume with six slices of 256 × 256) was used as support for both the super-pixel algorithm and network trained with simulated data (one-shot data), but no support was used for the GAN-trained model (zero-shot). This work used dice coefficient as a metric. The results demonstrate the better performance of the zero-shot, GAN-trained network on a real dataset against the one-shot network trained on real data using superpixel labels, with more than three times improvement in dice scores (50%–70% compared with less than 20%) obtained with the standard. Furthermore, this project tried to use one- and few-shot learning with the GAN-trained model, but the result did not improve as much.

This work also studied the segment anything foundation model published recently.[9] The team's preliminary analysis notes that the pretrained model fails to segment microstructures among other features and does not generalize well to the scientific imaging applications studied in this proposal. This project

[5] "Artificial Intelligence (AI)." DOE SC. Accessed February 15, 2024. https://science.osti.gov/ascr/Research/Artificial-Intelligence-AI
[6] A. Cheniour, A. Ziabari, and Y. Le Pape. "A Mesoscale 3D Model of Irradiated Concrete Informed via a 2.5 U-Net Semantic Segmentation." *Construction and Building Materials* 412, 2024. DOI: 10.1016/j.conbuildmat.2023.134392
[7] T. Park, A. Alexei, R. Efros, and J. Zhu. "Contrastive Learning for Unpaired Image-to-Image Translation." Proceedings, Part IX: Computer Vision—ECCV 2020: 16th European Conference, Glasgow, UK, August 23–28, 2020. DOI: 10.1007/978-3-030-58545-7_19
[8] C. Ouyang et al. "Self-Supervised Learning for Few-shot Medical Image Segmentation." IEEE Trans. On Medical Imaging, 2022. DOI: 10.1109/TMI/2022.3150682
[9] A. Kirillov et al. "Segment anything." arXiv preprint arXiv:2304.02643, 2023.

further tried to fine-tune the model with the used 42 slices of manually annotated data (each $1{,}024^2$) (that was used to train 2.5D U-Net[10]), but the improvement has been negligible.

In summary, this work observed that superpixel-based SSL was not viable for low-contrast data such as x-ray computed tomography of microstructures (more data exist but not shown here), but simulation- and GAN-based SSL drastically improve the performance. This result occurs despite the fact that no real data were seen during training of the latter two networks, and GAN-training data was also from a completely different scan than the test data. The proposed methods' results still need improvement especially when dealing with small pores and the boundary of microstructural features. This project also explored the generalizability of the segment anything foundational model and found it requires future fine-tuning with a large amount of labeled data to become useful for segmentation in scientific imaging applications.

[10] A. Cheniour, A. Ziabari, and Y. Le Pape. "A Mesoscale 3D Model of Irradiated Concrete Informed via a 2.5 U-Net Semantic Segmentation." Construction and Building Materials 412, 2024. DOI: 10.1016/j.conbuildmat.2023.134392

DEPUTY DIRECTOR DISCRETIONARY

10600: Cryogenic Ion Trap Engineering and Development

C. Seck, G. Buchs, J. Comish, J. Cui, C. Goetz, N. Peters, C. Redmon, K. Reed, G. Lynn, R. Pooser, W. Hicks III

Project Description

Simulating topological quantum materials systems could revolutionize quantum information science. Quantum simulators that harness qubits and quantum effects to emulate the Hamiltonians of open quantum systems are a key emerging technology that is well-suited for quantum topological system simulation. An ion trap–based cryogenic system is particularly well-suited for simulating quantum spin liquids, which is a key research area at ORNL in materials science and neutron scattering studies. Low temperatures (4 K) in these cryogenic ion trap systems increase qubit coherence by drastically reducing a broad array of noise sources. Additionally, careful engineering to reduce system vibrations significantly increases qubit coherence times and gate fidelities. These properties will enable quantum science research using higher-fidelity operations and larger qubit numbers, translating into accurate and efficient quantum simulations of realistic topological systems. The purpose of this project was to codesign (partnering with Infleqtion/ColdQuanta), acquire, and fine-tune a complete ultralow-vibration cryogenic apparatus that significantly improves upon state-of-the-art experimental systems.

Mission Relevance

When fully operational, the system will demonstrate sufficiently long multi-ion lifetime for calibrating and running algorithms, a significantly improved measured axial anomalous heating rate, and improved global 1- and 2-qubit gates owing to improved calibrations and lower heating rates. Such a system can serve as an integral part of beyond-exascale architectures and should benefit most programs and projects that can use quantum computing resources. Examples include protein folding calculations and vaccine development (biology applications), macromolecular properties calculations (materials science applications), and applications of interest in the environmental sciences.

Results and Accomplishments

Codesign, development, and on-site installation of the system was completed. The system is ready for characterization.

11261: Towards the 24th Core Capability

J. Newby, M. Demarteau, M. Febbraro, S. Castro Tognini, N. Novitzky, O. Hartbrich

Project Description

Exploiting existing and new facilities at the High Flux Isotope Reactor (HFIR) and Spallation Neutron Source (SNS), instrumentation was developed to study neutrino interactions in detail to reveal deviations from Standard Model predictions that could be indicative of the creation of a sterile neutrino, which is a hypothesized dark matter candidate. The second line of inquiry was the study to develop a new facility at SNS with higher sensitivity to study neutrino interactions in detail. Lastly, the possibility of fundamental physics experiments exploiting the excess beam power of SNS after the Proton Power Upgrade was examined.

Mission Relevance

The focus of this research is to expand the capability of ORNL-hosted user facilities toward a greater understanding of how the universe works at its most fundamental level. The neutrinos and other neutral particles produced at HFIR and SNS provide unique probes to measure the properties of neutrinos, study

their interactions with nuclei, and search for new candidates of matter. The lines of inquiry in this project relate to the objectives of DOE's HEP.

Results and Accomplishments

This project focused on expanding the neutrino experimental capabilities of ORNL's neutron user facilities on three fronts: a factor of 30 mass scaling of the liquid Ar instrument by developing large-scale evaporative coating techniques and an apparatus enabling low-threshold neutrino sensitivity at the ton scale, expanding the neutrino laboratory footprint at the First Target Station for multiton neutrino instruments, and the PROSPECT II neutrino instrument final design for HFIR.

Neutron background instruments were deployed at three First Target Station basement locations identified in FY 2022 as candidate neutrino locations compatible with SNS operations. These locations now have proton beam timing signals to enable further more-detailed feasibility studies.

The PROSPECT II instrument conceptual design was completed. The requirements document for the primary 4-ton liquid scintillator vessel was completed and submitted to the ORNL fabrication engineer for bidding and procurement.

The design and fabrication of the ton-scale liquid Ar instrument evaporative coating system was completed. This work enables the coating of 120 3-inch photomultiplier tubes and large Teflon reflector panels with tetraphenyl butadiene to shift the wavelength of photons emitted by Ar scintillation to longer wavelengths compatible with photon sensors.

A wavelength-shifting evaporative coating system for the fabrication of low-threshold, ton-scale neutrino and dark matter instrumentation was designed, fabricated, and commissioned. This new equipment will operate in a Building 6000 laboratory within a newly installed clean tent to fabricate the critical components for achieving low-threshold sensitivity to neutrino signals with the new 750 kg liquid Ar detector. Future experiments based on liquid Ar and other noble liquid technologies will benefit from this new capability.

10634: High Flux Isotope Reactor–Sustaining and Enhancing Neutron Science: Implementation and Integration

J. Busby, C. Bryan, D. Chandler, Z. Karriem, G. Hauck, D. Lockridge, D. Crawford, P. Mulligan, C. Goetz, Y. Zhang, M. Frost, D. Pointer, A. Campbell, J. McDuffee, G. Ehlers, B. Weston, B. Fuller, D. Chandler, T. Sowers

Project Description

The overall objective of this project was to provide a critical assessment of High Flux Isotope Reactor (HFIR) hardware, systems, and infrastructure that allow this unique facility to continue delivering world-leading scientific capabilities.

HFIR provides the world's highest neutron flux for critical missions in neutron scattering, isotope production, materials irradiation, neutron activation analysis, and nuclear physics. The ability to lead the world in each of these missions is unmatched, including high-performance reactors in Russia and France. To continue this record of excellence, an advisory committee for the DOE BESAC[1] published a report that provided guidance regarding national needs and priorities for reactor-based neutron sources in the United States.

This project detailed planning efforts for general scientific enhancements to continue, enhance, or introduce new, world-leading, scientific capabilities at HFIR. These scientific enhancements cover four major programmatic areas including (1) isotopes, (2) materials science, (3) physics, and

[1] R. Birgeneau et al. *The Scientific Justification for a US Domestic High-Performance Reactor-Based Research Facility.* Washington, DC: DOE SC BESAC, 2020. DOI: 10.2172/1647598

(4) nonproliferation research. Engaging with these programs, this project intended to conceptualize potential facility improvements for new, cutting-edge facilities that bring unique and world-leading capabilities to ORNL.

Mission Relevance

HFIR is a unique resource for the country. It is a key aspect of the nation's leadership in neutron scattering, isotope production, materials performance, nuclear fuels developments, and other science. Performing preconceptual work to further HFIR's capabilities and provide additional years of service benefits all programs and DOE.

Results and Accomplishments

This project captured and documented potential scientific improvements that could be realized at HFIR. Some of these concepts would require relatively small changes, and others are larger and could require new buildings or significant new equipment. Resulting from this work was a comprehensive list of 36 ideas, such as hot cell(s) and glove box location in the HFIR building and connected to the pool; a low-power critical facility; a larger gamma irradiation facility with instrumented access; an ion beam facility at HFIR; flexible flux trap configuration; experiment thimbles in the removable Be positions; and many others. Following the collection and documentation of ideas, the project pivoted to focused on the HFIR pressure vessel replacement and on determining which of the 36 concepts would benefit from being implemented during a HFIR pressure vessel replacement outage, as well as further definition of each of the concepts. This work is documented in a multivolume series of technical reports (ORNL/TM-2022/2691).

EMERGING AND CYBER SECURITY TECHNOLOGIES

This initiative did not have any completed projects in FY 2023.

ENZYME ENGINEERING

11048: High Throughput Polymer Characterization by Mass Spectrometry

J. Cahill, V. Kertesz

Project Description

This work aimed to (1) support routine, high-throughput characterization of soluble enzymatic products by using the PAL-drop/open-port sampling interface–mass spectrometry (OPSI-MS) technique; (2) evaluate methods to improve PAL-drop/OPSI-MS characterization, throughput, and sensitivity of enzymatic polymer products; and (3) develop a high-throughput thermal desorption–mass spectrometry (TD-MS) capability to characterize insoluble enzymatic products. This work addressed the initiative's priorities for incorporating high-throughput methods for characterizing soluble and insoluble products arising from enzymatic activity.

Mission Relevance

This work provided a scalable methodology (hundreds to millions of samples). Unique to the approach is the ability to measure soluble and insoluble oligomers with high throughput (<0.5–30 s per well) and to handle complex matrices such as cell lysates and different solvent compositions. High-throughput TD-MS capabilities are an innovative solution for examining the insoluble polymer products resulting from enzymatic reaction, with relevance to DOE BES and BER program objectives.

Results and Accomplishments

This work successfully scaled the enzyme characterization assay capabilities by developing an immediate drop-on-demand technology coupled with OPSI-MS. This capability exceeded throughput and sensitivity estimations. An approximately 3 s per sample throughput was achieved while using only 10 nL of analyte. This capability has the sensitivity to measure enzymatic nylon degradation products relevant to the initiative. A TD-MS capability was successfully built and tested. The results clearly indicate that the enzymes generate no insolubilized medium–molecular weight products.

11093: Mass Spectrometry Analysis for Platinum Enzyme Engineering

H. Andrews, B. Manard, V. Bradley, B. Sanders, A. Webb, M. Martin, Y. Ma

Project Description

Critical materials are key resources related to the manufacturing of clean energy technologies such as solar panels and electric vehicles. In response to the growing need for these materials, several research initiatives have evolved to diversify the supply chain and develop new ways to recycle or recover these elements. One of these initiatives is the Enzyme Engineering Initiative at ORNL. This initiative sets out to engineer enzymes with specific functionality, such as binding Pt-group elements. This engineering effort requires large volume throughput to design, build, and reiterate enzyme features. A keystone portion of this effort is benchmarking enzyme performance through analytical measurements. To evaluate metal binding performance, a novel analytical methodology—single-particle (SP)–inductively coupled plasma (ICP) mass spectrometry (MS)—was employed to investigate single magnetic beads with and without bound proteins to better understand Pt binding performance.

Mission Relevance

Critical materials refer to elements, typically rare earth elements or Pt group elements, which are key to continued advanced technologies. The DOE research agenda for critical materials focuses on cutting costs and improving environmental performance across the supply chain for the recovery of these materials. Creating new techniques for recycling critical materials is emphasized and could reduce the demand for

newly mined materials. This emphasis illustrates the growing interest in rapid analysis of Pt-group elements both because of the need to find new natural and engineered sources to satisfy increased demand in their use in various electronic devices and for strategic access to the US for new applications for these elements. Owing to their pivotal role in both military and civilian use, the dependable supply of these materials is crucial for both economic prosperity and national defense. This need makes the exceptional selectivity of the metal coordination environments identified in biological macromolecules an attractive alternative for the development of highly selective separation technologies for rare earth elements and Pt-group elements. Traditional analytical methods may provide inaccurate metal binding quantification due to the inability to distinguish bound and free Pt in samples. SP-ICP-time of flight (TOF)-MS resolves this issue by evaluating signals on a particle basis.

Results and Accomplishments

This project focused on developing and integrating new analytical approaches for benchmarking Pt binding of various mutant azurin-functionalized microparticles using SP-ICP-TOF-MS. Prior to working on samples for the enzyme engineering samples, the SP-ICP-TOF-MS system and its automated sampling system needed to be demonstrated and optimized. To do this optimization, a preliminary study on Au and Au/Ag nanoparticles (NPs) was performed.[1]

First, this project needed the ability to characterize Au nanoparticles of 50 and 100 nm, as well as 60 nm Ag-shelled Au core (Au/Ag) NPs, for their mass, respectively size, and isotopic composition in an automated and unattended fashion. In this work, an innovative autosampler was employed to mix and transport the blanks, standards, and samples into a high-efficiency SP introduction system for subsequent analysis by ICP-TOF-MS. Optimized NP transport efficiency into the ICP-TOF-MS was determined to be >80%. This combination—SP-ICP-TOF-MS—allowed for high-throughput sample analysis. In this project, approximately 50 total samples (including blanks or standards) were analyzed over 8 h to provide an accurate characterization of the NPs. This methodology was implemented over the course of 5 days to assess its long-term reproducibility. The in-run and day-to-day variation of sample transport was assessed to be 3.54% and 9.52% relative standard deviation, respectively. The determination of Au NP size and concentration was <5% relative difference from the certified values over these time periods. Isotopic characterization of the $^{107}Ag/^{109}Ag$ particles (n = 132,630) over the course of the measurements was determined to be 1.0788 ± 0.0030 with high accuracy (0.23% relative standard deviation) when compared with the multicollector–ICP-MS determination.

Following the NP study, an SP-ICP-TOF-MS procedure was developed to determine the Pt binding efficiency of protein-coated magnetic microparticles.[2] SP-ICP-TOF-MS is advantageous because of its ability to quasisimultaneously detect all isotopes (^{7}Li–^{242}Pu), allowing for both Pt and Fe (composition of magnetic microparticles) to be measured concurrently. This method subsequently allows for the differentiation between bound and unbound Pt. The 1 μm magnetic microparticles were fully characterized for their Fe concentration, particle concentration, and trace element composition by bulk digestion and SP-ICP-TOF-MS. The results of both approaches agreed with the certificate values. Using the single-particle methodology, the Pt loading was quantified to be to 0.18 fg/particle and 0.32 fg/particle for the streptavidin-coated and azurin-coated microparticles, respectively. Both streptavidin-coated and azurin-coated microparticles had a particle–Pt association >65%. The Pt-bound samples were also analyzed via bulk digestion-based ICP-MS, and the results agreed when the particle–Pt association rates determined from single-particle analysis were applied to account for unbound Pt in the samples. This result highlights the importance of single-particle analysis for closer inspection of Pt binding performance. The SP-ICP-TOF-MS approach offers advantages over typical bulk digestion methods by eliminating laborious sample preparation and enabling differentiation between bound and unbound Pt in a

[1] B. T. Manard et al. "Towards Automated and High-Throughput Quantitative Sizing and Isotopic Analysis of Nanoparticles via Single Particle-ICP-TOF-MS." *Nanomaterials* 13.8, 2023, 1322. DOI: 10.3390/nano13081322. Highlighted on journal cover.
[2] H. B. Andrews et al. "Characterizing Platinum Binding on Protein-Functionalized Magnetic Microparticles using Single Particle-ICP-TOF-MS." Submitted to *Analytical Chemistry*, 2023.

solution and quantification of Pt on a particle-by-particle basis. The procedure presented here enables quantification of metal content per particle, which could be broadly implemented for other single-particle applications.

11114: Medium Throughput Polymer Characterization Using Raman Spectroscopy, Differential Scanning Calorimetry, and Broadband Dielectric Spectroscopy

V. Bocharova, C. Gainaru, A. Sokolov

Project Description

Enzymes play a crucial role in polymer upcycling; however, current analytical methods for quantifying the efficiency of enzymatic reactions are relatively crude and slow. Addressing these limitations will facilitate the rapid development of innovative biochemical solutions to various challenges in materials science. This project aimed to establish an analytical experimental platform that can collect key polymer characteristics via comprehensive and accelerated characterization of enzymatic hydrolysis reaction products and reaction kinetics. The approach provides a set of unique experimental techniques that can address the complex physicochemical characteristics of soluble and insoluble products of enzymatic hydrolysis reactions, establishing a connection between polymer properties and enzyme structure and activity. The experimental techniques assessed changes in crystalline structure by using differential scanning calorimetry methods to compare melting enthalpies of the initial and modified polymer, molecular dynamics via broadband dielectric spectroscopy, and molecular weight via gel permeation chromatography. The results of this work enhanced the understanding of the mechanisms underlying enzyme functioning in polymer upcycling, ultimately leading to the accelerated discovery and design of new functional enzymes.

Mission Relevance

Enzymes excel at performing challenging chemical transformations with high specificity under benign conditions, making them ideal candidates for the biodegradation of plastics. The activity, substrate specificity, and stability of natural enzymes can be further enhanced by enzyme engineering—a research area covered by DOE's BER and BETO programs. These programs can address the environmental challenge critical for the DOE mission of ensuring US security and prosperity and the ORNL mission of delivering scientific discoveries that accelerate the development and deployment of solutions in recycling while creating economic opportunities for the nation.

Results and Accomplishments

The team established a workflow using a set of selected experimental tools to characterize the hydrolysis of polyamides. This approach can be applied to characterize the reaction kinetics and understand the mechanism of enzymatic action for any new and unknown hydrolyzing enzyme. The workflow was tested with a known enzyme, revealing a new aspect of the enzymatic mechanism concerning the surface area of the substrate. This work generated benchmark polymer degradation data from a known hydrolase enzyme sample by using differential scanning calorimetry, thermal gravimetric analysis, and gel permeation chromatography. The results of this work are included in a paper that is currently under preparation.

11160: High-Throughput Protein Stability Assays: Mutation and Binding

A. Sedova, O. Demerdash

Project Description

The overarching goal of this project was to determine how mutations affect protein thermal stability and ligand binding using high-throughput experimental methods and was assisted by advanced computational

approaches and data analytics. To accomplish this goal, the project aimed to develop the protein thermal stability/shift assay pipeline together with computationally assisted interpretation using high-performance computing simulation to aid data analysis for both high-throughput stability evaluation and binding affinity measurements on mutated proteins. It focused on mutations that alter the binding of elemental Pt and Pt-containing species by proteins for selective separation from feedstocks and waste streams. These efforts will provide high-accuracy thermodynamic and kinetic data for understanding the role of mutations in binding, benchmarking high-throughput methods, and providing data for learning and simulation efforts.

Mission Relevance

The connection between design, testing, and learning is enabled and enhanced by new assay capabilities, automation, large datasets for learning and statistics, high-throughput experiments, and accurate data analysis methods that make use of ORNL's high-performance computing resources. Energy critical materials and their extraction and reuse from limited supply streams is an important focus in the push for energy innovations and clear energy solutions.

INTEGRATED STUDIES OF COMPLEX BIOLOGICAL AND ENVIRONMENTAL SYSTEMS

10305: Linking Genes to Ecosystems with Phytoliths

N. Griffiths, X. Yang, M. Martin, W. Muchero, E. Herndon, H. Andrews, M. McLennan, H. Li, S. Shelley, A. Wymore, F. Santos, V. Salmon, B. Wang, A. Carrell, A. Webb, G. Schwaner, S. Martin

Project Description

The element Si enhances plant tolerance to various stressors, and the accumulation of Si in C-containing phytoliths can have ecosystem-level consequences by contributing to long-term C sequestration. However, the accumulation of Si in plant tissues inhibits bioprocessing applications from bioenergy feedstocks. Genetic variability and molecular mechanisms of Si accumulation in bioenergy crops are unknown, and few studies examine the consequences of genetic variability on ecosystem-level processes. Therefore, the goal of this research was to identify genes underlying Si accumulation in bioenergy crops and to use that knowledge to modify Si accumulation and evaluate leaf decomposition rate. This project used a genome-wide association study (GWAS), facilitated by high-throughput examination of Si accumulation using laser-induced breakdown spectroscopy (LIBS), as well as translational genomics, to identify candidate genes associated with Si accumulation in *Populus* (poplar). Then, this study created transgenic poplar plants that overexpress multiple combinations of candidate genes for Si accumulation. The project also quantified Si accumulation of different poplar genotypes that expressed high vs. low levels of Si transporter genes using scanning electron microscopy and energy dispersive spectroscopy. Lastly, this work assessed ecosystem-level effects with field-based decomposition experiments.

Mission Relevance

By demonstrating how genetic alterations can affect ecosystem processes—and by verifying a complete genes-to-ecosystem approach—this research strongly aligns with the upcoming DOE Genes to Ecosystem Initiative. The capabilities developed from this research address the objectives of DOE BER programs, including Bioenergy Research Centers, Biosystems Design, and Plant Science for Bioenergy.

Results and Accomplishments

By integrating ORNL's capabilities in quantitative genetics, synthetic biology, plant phenomics, environmental chemistry, and ecosystem ecology, this collaborative project developed a robust approach for evaluating how genetic alterations may affect ecosystem processes. The goal of this research was to identify genes' underlying Si accumulation in bioenergy crops (poplar) and to use that knowledge to modify Si accumulation in poplar and evaluate C sequestration potential. To achieve this goal, this project was structured around three interrelated tasks: (1) identify the genes associated with Si accumulation in poplar, (2) engineer Si transporter genes in poplar, and (3) characterize poplar plants for Si accumulation and decomposition rate.

Methods and results relating to initiative identification of Si accumulation genes in poplar is published in *Plant Biotechnology Reports*.[1] Following that work, LIBS was used to evaluate the Si levels in woody poplar tissue. The woody poplar tissues were pelletized (n = 220 total pellets), and then the pellet surfaces were scanned to collect an average spectrum for each genotype. Additionally, a sample with no detectable Si was spiked with a Si solution to form a set of matrix-matched calibration standards. The resulting calibration curve was used to quantify the Si levels in the tissues, and the results were used as input for the GWAS. The GWAS identified genotypes that appeared to be high or low expressors of the Si transporter genes, and these genotypes were used for characterization studies (Task 3). The four Si

[1] M. M. Hassan et al. "Genome-Wide Identification and Functional Prediction of Silicon (Si) Transporters in Poplar (Populus trichocarpa)." *Plant Biotechnology Reports* 17, 2023, 285–302. DOI: 10.1007/s11816-022-00788-4

transporter genes identified in this task were BESC-152, BESC-315, CHWH-27-5, and GW-11053. The two BESC genes were low expressors, and the other two were high expressors.

A synthetic biology approach was used to create transgenic poplar plants that overexpress multiple combinations of candidate genes for Si accumulation. Transgenic plants engineered with Si transporter genes were confirmed by an eGFPuv signal. Following treatment with a 1.7 mM Si–water solution for 10 days, the average concentration (represented by LIBS emission intensity) of Si in PtSi-line2 plants was almost twice that in wild-type plants. These results indicate that overexpression of endogenous Si transporter genes in poplar can significantly enhance Si accumulation.

Phytolith characterization and quantification were completed on intact leaves from 13 individual *P. trichocarpa* trees from two common gardens—4 of the 13 were from Clatskanie, Oregon, and 9 were from Davis, California. These individuals represented four genotypes with different expected Si uptake rates based on their rates of expression of genes associated with Si uptake. Using environmental scanning electron microscope energy dispersive x-ray spectroscopy, the study determined that genotype poorly predicted leaf Si concentrations because trends were either unclear or opposite of the hypothesized trends. From these findings, this project concluded that environmental factors and possibly phenology control Si uptake more strongly than genotype.

The team hypothesized that leaves with higher Si content may have more organic C occluded within Si-rich phytoliths, which would result in slower decomposition rates. However, the differences in Si content did not translate into differences in decomposition rates. In fact, there appeared to be no clear effect of intrinsic nor extrinsic drivers on decomposition rate. The final Si content of decomposed leaves was not correlated with mass remaining.

10320: Defining Microbial Gene Function in Complex Communities

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Project Description

This project focused on developing methods to measure how the interactions between organisms in a complex community influence the physiology and function of its members. Most of what scientists know about microbial gene functions comes from studying microbes in monocultures in a laboratory environment, yet many genes are not expressed under these conditions. This project aimed to develop a robust strategy to identify interaction-induced phenotypes, including growth and metabolite production, and to characterize the molecular mechanisms underpinning these phenotypes using analytical technologies. The project had the goal of assigning new or expanded functions to microbial genes. To reach this goal, researchers cocultured microbes in pairwise combinations in a multiple–well plate format and used optical and chemical imaging to identify differences in growth rates and metabolite production in cocultures compared with monocultures. For select cocultures, researchers then performed proteomics to identify candidate gene products that were differentially abundant, suggesting that they might contribute to the observed phenotypes. Within this study, researchers generated highly correlated datasets on microbial interactions, growth patterns, and metabolite profiles; these datasets can be used to drive hypothesis generation and ultimately feed larger-scale predictive models for soil- and plant-associated communities.

Mission Relevance

This project developed methods to rapidly characterize interaction-specific growth patterns and metabolites in mixed bacterial communities using optical and chemical imaging. The project aimed to assign new and expanded functions to microbial genes. This new technology can be applied to DOE-relevant biosystems to generate high-content data for systems biology and predictive modeling to increase and enhance knowledge about complex biosystems.

Results and Accomplishments

The initial work on this project involved developing and optimizing the individual optical and chemical imaging experimental workflows to allow multimodal imaging. The workflows were then combined to enable the collection of correlated optical and chemical imaging datasets from bacterial cocultures. During this project, researchers collected optical imaging datasets for hundreds of bacterial cocultures and correlated optical and chemical imaging datasets from at least 12 coculture pairs. Based on these data, researchers performed a deeper analysis of the coculture of *Pantoea* YR343 and *Flavobacterium* CF108, which showed growth phenotypes and the production of several new metabolites compared with the single strains. Using tandem mass spectrometry, this project identified metabolites with the empirical formulas of $C_{14}H_{16}N_2O$ and $C_{14}H_{17}N_3O$. Researchers were also able to determine that an additional metabolite is a cyclodipeptide: cyclo(phenylalanine proline).

The project then examined how the proteomic profiles of *Pantoea* YR343 and *Flavobacterium* CF108 differed when they were grown in coculture or in a monoculture. Based on protein abundance, this study found that the *Flavobacterium* CF108 comprised 18% of the coculture, and *Pantoea* YR343 accounted for 82%, which is consistent with the optical imaging data. The proteomic data indicated that the *Pantoea* strain showed upregulation of genes involved in exopolysaccharide biosynthesis, downregulation of flagellar genes, and regulation of genes involved in cyclic dimeric GMP signaling, which are all consistent with a biofilm lifestyle. In *Flavobacterium* CF108, this study observed upregulation of gliding motility and carbohydrate metabolism.

Researchers also looked specifically at proteins encoded by biosynthetic gene clusters in both strains with the hope of identifying pathways that might be involved in the production of the identified metabolites. In *Flavobacterium* CF108, this study found evidence of a gene cluster that might be responsible for the production of cyclo(phenylalanine proline), and current work is testing that hypothesis by constructing a knockout strain using genetic engineering. As researchers developed and tested this new capability, they discovered a bottleneck in throughput resulting from the more labor-intensive chemical imaging data collection and analyses. To counter this issue, this study added a prescreening step in the workflow to focus the chemical imaging efforts on cocultures that are likely to show interaction-induced phenotypes by monitoring the optical density and fluorescence of cocultures grown in liquid media in 96-well plates using one fluorescently tagged bacterial strain in combination with different untagged bacterial strains. Wells that showed significantly higher or lower fluorescence levels compared with the control strains (i.e., the fluorescent bacteria alone) were flagged as candidate cocultures of interest. Researchers then collected the supernatant from these cocultures and examined metabolite profiles using a high-throughput PAL-drop instrument. The cocultures whose metabolite profiles differed from those found in monocultures were then subjected to the full optical and chemical imaging workflow, as originally developed.

10718: Phenotyping and Predictive Analytics Using Hyperspectral Imaging

S. Martin, H.-J. Yoon, B. Christian

Project Description

This project aimed to create a hyperspectral image analysis pipeline based on imagery from the Advanced Plant Phenotyping Laboratory (APPL), SpecimIQ, and other hyperspectral instrumentation. This work used cloud native compute infrastructure provided by ORNL to conceive, develop, test, and deploy the software described in the project's final report.

Mission Relevance

Hyperspectral imagery holds promise as a robust tool for rapid phenotyping of plants. The APPL facility and other instrumentation has the capability to collect hyperspectral data. However, deep analytics and

trained datasets are needed for this technology to reach its full potential. This project was a first step in enabling analytics across a spectrum of instrumentation available to researchers through the APPL facility.

Results and Accomplishments

A pipeline was conceived, developed, and deployed on the Oak Ridge Leadership Computing Facility's cloud native infrastructure to analyze imagery from the APPL facility. A full description of the project and its algorithms has been published.[2] A corresponding software package has been released.[3]

This project represents one of the first tangible examples of cloud native computing for production biological systems undertaken at ORNL. In successfully completing this project, the team has created a road map for cloud native computing in the biology/life sciences space. In addition to the scientific value of the project, the lessons learned relating to cloud native data management, computing, resource allocations, and user interfaces are directly applicable to other similar projects.

10844: Ecosystem Resilience to Thermal Extremes: Urbanization Impacts

J. Mao, D. Ricciuto, X. Shi, D. Rastogi, C. Brelsford

Project Description

Because of the complex interplay of urbanization and climate, the patterns of urban ecosystems are highly dynamic. Accurately understanding the responses of urban ecosystems to their surrounding environments, especially the intensified thermal extremes, is challenging. This project assembled and analyzed empirical data to quantify the resilience of selected ecosystem processes to major extreme thermal perturbations (e.g., heat waves) across representative US cities. The project performed advanced statistical analyses to determine significant urban and rural differences in resilience, thresholds, and underlying mechanisms. The project also identified and optimized the most relevant parameters to urban ecosystem resilience in the land component of the DOE's Energy Exascale Earth System Model (E3SM), called the E3SM Land Model (ELM), and quantified the extent to which ELM performance on urban ecosystem resilience can be improved through parameterization. The optimized version of ELM and downscaled environmental drivers were also used to conduct test simulations at high spatiotemporal resolutions (1 km every 3 h) in the twenty-first century. Using the projected model results, this project determined when, where, and which urban ecosystem processes may be most or least resilient to simulated future thermal extremes.

Mission Relevance

This project's mechanistic analysis and modeling framework aligns with this initiative's strategy by advancing the understanding of understudied urban ecosystems' resilience to extreme thermal perturbations. It leveraged existing ORNL capabilities on field observations and manipulations, ELM diagnostics and development, climate downscaling, and E3SM benchmarking and parameter uncertainty quantification. New mechanistic understanding, modeling capabilities, and future high-resolution resilience sensitivity simulations will inform the new DOE BER Urban Integrated Field Laboratories, providing a foundation for urban site selection and key thermal perturbation experiments. This project also falls under the scope of "advance modeling and understanding of important ecological, biological, and carbon cycle interactions and feedbacks in the climate system to identify potential tipping points and

[2] H.-J. Yoon et al. *HuberKube: A Kubernetes Based System for the Automation of Processing and Analysis of Hyperspectral Data Obtained from Multiple Hyperspectral Imaging Systems.* ORNL/TM-2024/3279. Oak Ridge, Tennessee: Oak Ridge National Laboratory, 2024. DOI: 10.2172/2305378
[3] H.-J. Yoon and S. Martin. *HyperKube*. Oak Ridge, Tennessee: Oak Ridge National Laboratory, 2023. DOI: 10.11578/dc.20231101.1

possible energy strategies" from the BERAC,[4] as well as "enhance efforts to ensure that the results of experiments and observations are used to inform model development and, conversely, that model outputs and simulations are used to guide the design of field experiments and new observational efforts" from DOE.[5]

Results and Accomplishments

With continuing global warming and urbanization, a better understanding of the resilience of urban vegetation to heat waves is valuable for maintaining urban ecosystem services and human well-being, but few studies have addressed this topic at a large scale. The team quantified and compared the responses of urban and rural vegetation with heat waves during 2001–2019 for 85 major cities across the contiguous US using monthly Enhanced Vegetation Index (EVI) data and two metrics (*resistance*, the relative difference between EVI during the heat wave and pre-event EVI, and *recovery*, the relative difference between postevent EVI and pre-event EVI). This work found consistent patterns of urban–rural differences in both metrics. In winter, the urban areas had more positive metrics than the rural areas. In the other seasons, the urban areas had more positive metrics in the western US but more negative metrics in the eastern US. The east–west difference can be explained by the considerably higher optimal growth temperatures and lower water stress of urban vegetation compared with rural vegetation in the western US. The magnitude of the metrics was generally smaller in urban areas than in rural areas, with the greatest difference occurring where the rural areas are dominated by deciduous forests and the least difference occurring for rural evergreen forests. Further analysis at a 1 km pixel level showed that impervious fraction, the fraction of the rural vegetation cover, local urban heat island intensity, and water stress were the key factors giving rise to the city-scale urban–rural differences. Overall, the findings show that urban–rural differences in thermal, water, and land cover environment were the key drivers of vegetation resilience to heat waves.

These findings contribute to scientific knowledge by showing that temperature, moisture, vegetation type, and impervious area factors were important to both large- and small-scale variations in vegetation resilience to heat waves. The findings have implications for urban forestry and ecosystem management. For example, the delayed negative responses suggest that monitoring vegetation health is important after heat waves and that cost-effective prevention of negative effects from heat waves may be achieved by focusing on the longest and most intense heat waves. The importance of moisture conditions suggests that promoting urban vegetation growth requires not only managing the urban heat island but also the urban dry/wet island. The mechanisms underlying the adaptation of urban vegetation to the warmer urban environment through higher optimal temperatures, as well as the effects of land cover types and impervious area on urban vegetation resilience, require future studies to clarify. In this regard, urban foresters and ecosystem managers are uniquely positioned in their access to detailed municipal-level data (e.g., tree species, lidar maps, soil surveys, air quality stations). Such data will help them understand ground changes during heat waves that cannot be resolved by remote sensing information over large regions, make better decisions such as selecting tree species and issuing land development permits, and better quantify ecosystem services in relation to remotely sensed vegetation changes. These findings advance the understanding of the effects of urbanization on vegetation growth and suggest that urban planners and ecosystem managers should focus on those key drivers to promote urban vegetation resilience to heat waves. Relevant research has been developed into a paper, which is under review by the *Proceedings of the National Academy of Sciences.*

In addition to the previously highlighted mechanistic analyses, the project introduced new capabilities. Specifically, the team developed high-resolution ELM simulation workflows involving the preparation of detailed land property and meteorological forcing datasets, targeted ELM runs for urban regions, and

[4] BERAC Subcommittee on Grand Research Challenges for Biological and Environmental Research. *Grand Challenges for Biological and Environmental Research: Progress and Future Vision 2017.* Washington, DC: DOE BERAC, 2017.
[5] DOE SC BER. *Earth and Environmental Systems Sciences Division Strategic Plan*. Washington, DC: DOE, 2018.

postprocessing of relevant ELM simulations. Extensive test simulations, using various drivers and model parameters, were conducted in Chicago, Illinois, for both historical and future periods. Ongoing efforts involve thorough simulation analysis and comparisons with observations. Furthermore, the project established a framework for testing and calibrating ELM in urban sites using the offline land model test bed. Leveraging leaf area index, gross primary productivity, and evapotranspiration observations, the project successfully calibrated five model parameters for three plant functional types across selected urban and rural grids in different cities. The optimization of ELM and subsequent simulations were instrumental in enhancing the projection of future ecosystem resilience.

MATERIALS INNOVATION: DIGITAL METALLURGY

10222: Operando Neutron Characterization of Metal Additive Manufacturing

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Project Description

This project delivered a new capability to ORNL and the scientific community as a whole for operando neutron characterization of metal additive manufacturing (AM). Through the development and integration of this research platform into the VULCAN beamline at the Spallation Neutron Source (SNS), this work sought to achieve three key aims: (1) understand nonequilibrium, in situ phase transformation phenomena in structural materials during advanced manufacturing; (2) determine thermal stress development during processing and its relationships to microstructure, phase transformations, and complex boundary conditions; and (3) apply artificial intelligence techniques to accelerate neutron data acquisition and analysis. To achieve these aims, this project used low-temperature transformation (LTT) steel as a model material system that exemplifies the effect of operando neutron data to quantify the relationships between phase transformations and stress evolution. This project constructed the AM platform using a wire-arc deposition process and demonstrated its functionality. The system was integrated into the VULCAN beamline, and the team demonstrated artificial intelligence–accelerated operando neutron data collection for phase transformations and thermal stress in an LTT steel. Additionally, a laser deposition head for use with wire or powder feedstock was integrated into the device.

Mission Relevance

This project has the potential to enable significant scientific advances in structural materials and advanced manufacturing. It creates a unique capability to study structural materials during manufacturing and demonstrates the effects of neutron science on the future of manufacturing research. These capabilities enable ORNL to fulfill its mission by designing new materials and advanced manufacturing practices to address critical challenges in clean energy, energy efficiency, and national security.

Results and Accomplishments

The purpose of this project was to design, fabricate, and commission an AM system for performing operando neutron diffraction studies. The selected material for this project was an LTT steel, the thermomechanical properties of which were first characterized in detail.[1] The system was then commissioned at the VULCAN beamline at SNS through a series of experiments using this material to demonstrate successful processing and neutron data collection and analysis. OpeN-AM was developed and commissioned with time-resolved diffraction data collected during processing of an LTT steel.

Aside from enabling the collection of operando neutron diffraction data, a number of other innovations were developed to maximize the scientific impact of experiments using the system. A custom sensing system was implemented, which used multiple high-resolution optical cameras and hyperspectral infrared imaging. These data were successfully registered with the neutron data to enable correlated data analysis. Additionally, a software tool was developed to perform dynamic samples of mapped neutron diffraction data by streaming data in real time and using it to update a Gaussian process model. The model was then used to select the most valuable data points for subsequent measurements, reducing the total data

[1] W. Tang et al. "Temperature-Dependent Thermal and Mechanical Properties of a Wire Arc Additively Manufactured Low Transformation Temperature Steel." *Metall. Mater. Trans. A* 54, 2023, 854–868. DOI: 10.1007/s11661-022-06933-6

collection time by up to 60% for equivalent accuracy of results. This software has been successfully implemented at both SNS[2] and High Flux Isotope Reactor beamlines.[3]

The collection of these capabilities was used to gain a scientific understanding of the strain evolution in an additively manufactured LTT steel component. The neutron diffraction data showed trends in elastic lattice strain, plastic strain accumulation, and annealing as a function of the temperature histories and resulting phase transformations in the LTT steel. These data were further correlated with infrared imaging and used to calibrate and validate a computational model. These results were reported in a manuscript published in *Nature Communications*.[4]

10838: Metastability-Driven Design of Additive Manufacturing Aluminum Superalloys

Y. Yang, Y. Osetskiy, K. An, R. Dehoff, W. Zhong, A. Plotkowski, A. Perrin, Y. Lin, Q. Q. Ren, E. George

Project Description

The $L1_2$-type of Al_3M (M = Ti, Zr, Hf, V, Nb, or Ta) phases are attractive options for use as strengthening phases in Al alloys or as a basis of a single-phase alloy. However, introducing a high-volume fraction of $L1_2$_Al_3M precipitates is difficult because of limited solubility of metal in the Al face-centered cubic structure. Even more challenging is that the Al_3M usually forms complex crystal structures (such as tetragonal DO_{22}/DO_{23}) instead of the simple $L1_2$ structure, which causes a large lattice misfit along the C-axis direction and inferior coarsening resistance. The complex crystal structures also make the single-phase Al_3M alloys more brittle because of a lack of independent slip systems. Additive manufacturing (AM) offers opportunities to produce intentional metastable microstructures with extensive solute trapping and simple crystal structures because of high cooling rates during the AM process. The relationships between process parameters, heat transfer, and metastable microstructure, however, are often convoluted and poorly understood. The ability to understand and control these metastable phenomena has the potential to revolutionize the design of materials and microstructures that cannot be achieved through conventional casting methods.

This project experimentally screened the relative phase stability of stable structure (DO_{22}/DO_{23}) and metastable structure ($L1_2$) in six binary Al–M alloys (M = Ti, Zr, Hf, V, Nb, and Ta). Theoretic modeling was performed to understand the thermodynamics and kinetics that control competition between the nucleation and growth of stable and metastable phases. Based on screening results, Al–Zr and Al–Nb binary alloys were selected for AM (laser beam powder bed technique). The as-build Al–Zr and Al–Nb alloys were then subjected to in situ tensile testing under neutron diffraction. The results showed that the Al–Zr alloy predominantly consists of the metastable $L1_2$ phase, and the Al–Nb alloy mainly contains the stable DO_{22} phase. Both alloys show a favorable combination of strength and ductility. This work demonstrated the feasibility to use a metastable phase for strengthening the AM alloy design. This work also found that the size and morphology of the strengthening phase is more critical than the crystal structure in determining the mechanical behavior of AM Al alloys. This work opens a new avenue to design high-strength, high-ductility Al alloys for AM applications to the transportation, aerospace, and defense industries.

[2] S. V. Venkatakrishnan et al. "Adaptive Sampling for Accelerating Neutron Diffraction-Based Strain Mapping." *Mach. Learn. Sci. Technol.* 4, 2023, 025001. DOI: 10.1088/2632-2153/acc512
[3] C. M. Fancher et al. "Validating the Use of Gaussian Process Regression for Adaptive Mapping of Residual Stress Fields." *Materials* 16, 10, 2023, 3854. DOI: 10.3390/ma16103854
[4] A. Plotkowski et al. "Operando Neutron Diffraction Reveals Mechanisms for Controlled Strain Evolution in 3D Printing." *Nat. Commun*. 14, 2023, 4950. DOI: 10.1038/s41467-023-40456-x

Mission Relevance

This study used ORNL's science-based computational and experimental capabilities to establish a knowledge base on synergy between the metastability of phases and rapid/heterogeneous cooling conditions in AM. The proposed $L1_2$ precipitate–strengthened, Al-based superalloys is part of a class of lightweight, high-temperature, high-strength Al alloys that has applications in energy-efficient transportation sectors. The framework of the innovative AM Al alloy design in this study can also be used for other systems with the similar challenges.

Results and Accomplishments

Six binary alloys—Al–2 atom % M (M = Ti, Zr, Hf, V, Nb, and Ta)—were made through arc-melting. These alloys were then subjected to laser remelting. Although only the stable DO_{23} in Al–Zr and DO_{22} in Al–Nb precipitates were observed in the as–arc-melted structure, metastable $L1_2$ precipitates were observed in the laser-remelted regions, suggesting that a fast cooling rate does promote the formation of the metastable L12 structure, which is consistent with theoretic modeling. The precipitate phase in the as-cast region was coarse and faceted in Al–Zr alloy, which usually leads to inferior mechanical properties. However, under the laser remelting condition, the precipitate phase showed a much more refined and equiaxial morphology and was predominantly identified as $L1_2$. On the other hand, the precipitate phase in the Al–Nb alloy showed primarily the DO_{22} phase in the as-cast region. These precipitates became more refined after laser remelting, but their crystal structure remained primarily as DO_{22}.

Based on the microstructure screening results in MS3, the Al–Zr and Al–Nb alloy systems were selected for bulk AM. The as-printed Al–2Zr and Al–2Nb alloys showed good printability and had a minimal number of defects, such as porosity and cracks. These as-printed Al–Zr and Al–Nb alloys were then subjected to in situ tensile testing under neutron diffraction. The results showed that the volume fraction of $L1_2$ 7.44% is close to the predicted value (7.42%), suggesting the as-printed Al–Zr alloy has a composition close to the nominal value. In contrast, the Al–Nb alloy only had a volume fraction of 2.69% DO_{22}, which is significantly less than the theoretical calculation of 7.11%, suggesting significant loss of Nb in as-printed alloys. The stress–strain curve showed a good combination of strength (ultimate tensile strength of 300 MPa) and ductility (total elongation of 9%) in the as-built Al–Zr alloy. Given the low volume fraction of the precipitate phase in the Al–Nb alloy, the strength (ultimate tensile strength of 200 Mpa) is lower than that of the Al–Zr alloy. The load sharing curves suggested that both DO_{22} and $L1_2$ serve as good strengthening phases. The Al–Nb alloy, if it had reached the nominal composition, was anticipated to display a similar high strength to that of the Al–Zr alloy despite DO_{22} having a lower symmetry than the $L1_2$ in the Al–Zr alloy. These results suggest that the precipitate morphology, size, and distribution are more critical factors for the mechanical behavior than the crystal structure in these alloys.

10881: Transformational Approach to Digitally Manufacture Structural Alloys with Co-Optimized Strength and Environmental Resistance

R. Pillai, R. Kurfess, S. Nag, Y.-F. Su, Q. Q. Ren, B. Fillingim, T. Feldhausen, T. Lowe, B. Johnston, A. Willoughby, K. Epps, J. Moser

Project Description

A paradigm shift in the traditional sequential design approaches is critically essential to create application-specific, hierarchical, and multifunctional materials with superior long-term performance for next-generation energy technologies involving extreme environments. In this research, this project aimed to leverage the flexibility and geometric/compositional complexity offered by additive manufacturing to demonstrate this new approach by codesigning a compositionally graded, Ni-based structural alloy (no coatings) to enable mitigation of the environmental degradation of surfaces exposed to molten halide salts while simultaneous suppressing the consequent deterioration in mechanical stability. Thermokinetic modeling describing the underlying physics of thermally and environmentally induced spatiotemporal,

compositional, and microstructural evolution were employed to predict the parameter space of material deposition processes and precisely identify the required composition gradient. Essentially, the thermodynamic driving forces for species transport will be designed after accounting for corrosion-induced surface degradation during service. This unique combined computational–manufacturing technique was validated by evaluating the performance of the resultant fabricated material in realistic severe operating environments.

Mission Relevance

This research is directly aligned with the objective of developing materials for extreme environments that cannot be manufactured with conventional approaches and enable simultaneous, application-specific development of materials. It supports DOE's mission by improving the understanding of physical phenomena and to develop advanced materials and processes to enable energy-efficient, cost-competitive, and environmentally acceptable materials technologies for a variety of important national priorities.

Results and Accomplishments

Various compositional gradients between the terminal chemistries of Hastelloy-N (HN) and Haynes 282 (282) were calculated with a coupled thermodynamic–kinetic model. The choice of the compositional grading will strongly depend on the desired mechanical strength, which will be determined through mechanical testing of graded material specimens. None of the chosen compositional gradients demonstrated formation of detrimental brittle phases and indicate very low susceptibility to cracking during fabrication. Additionally, diffusion couples of HN and 282 were made to evaluate the chemical and structural compatibility between the two terminal chemistries. Characterization of the annealed and heat-treated specimens machined from the diffusion couple did not show the presence of any brittle phase formation.

Next, sample builds of terminal alloy chemistries with <20 vol % of brittle intermetallic phases were generated. The terminal chemistries of HN and 282 were fabricated with blown powder directed energy deposition. Detailed characterization of the as-deposited microstructure showed the presence of the phases typically present in the wrought variants: γ-face-centered cubic (fcc) + γ′-fcc + Mo-rich (M_6C-type) carbides for HN and γ-fcc + Ti-rich (MC-type) carbides for 282. No brittle intermetallic phases were observed.

The initial cubic (2 × 2 × 2 mm) build of the dual material HN–282 (i.e., HN on 282 without compositional grading) was fabricated. Mechanical testing of the dual HN–282 material was completed. Tensile and creep specimens were machined from linearly and radially graded HN–282 material. Tensile testing was performed at room and elevated temperatures. Creep testing was performed at 760°C (336 and 290 Mpa) and at 816°C (173 Mpa).

The dual material has performed significantly better than HN with almost a 200% increase in the yield strength (YS) and 140% increase in the ultimate tensile strength (UTS). The strength of the dual material was lower than 282, but the obtained values for YS and UTS are considerably higher (150% for YS and UTS) than the solid solution-strengthened alloy 230, which is a candidate material for heat exchangers in concentrating solar power applications and is highly susceptible to molten salts corrosion.

Additionally, creep testing of the terminal chemistry 282 in the binary molten $NaCl–MgCl_2$ salt and in air was conducted to evaluate the effect of molten salt corrosion on the creep behavior of 282. A 15% reduction in time to rupture, 25% reduction in time to 2% creep strain, and five times higher creep strains were observed for the specimen exposed to the molten chloride salt.

To demonstrate the applicability of the approach to a broader range of materials, the nuclear code–qualified austenitic steel 316H was fabricated with directed energy deposition. 316H is known to be susceptible to molten salt corrosion, and creep testing performed in molten salts showed a significant debit in creep rupture lifetimes for 316H. The fabrication of an HN–316H dual material was undertaken

and completed. This material solution is expected to be of interest for molten salt reactor components, with 316H being the preferred structural material for these applications.

This project demonstrated the applicability of diffusion couples to guide the fabrication of graded materials, which can enable a significant reduction in developmental effort and provide valuable insights into the optimization of property-graded materials. The project established a unique experimental–computational–manufacturing framework at ORNL that will allow fabrication of property-graded, high-temperature alloys with co-optimized environmental resistance (dual-corrosion atmospheres) for operations in harsh environments. The success of this approach can ultimately eliminate the need for corrosion-resistant coatings and provide a unique pathway to design high-temperature alloys for extreme environments.

10894: Thermodynamic Design of Multi-Material Additively Manufactured Structures

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Project Description

The goal of this research was to develop additive manufacturing (AM) approaches and processes to enable the fabrication of steel–Al joints that are significantly more robust than is possible with conventional joining techniques. Joining steel and Al is a critical technology for many applications, including for reducing structural weight, but fabricating these joints with conventional techniques typically leads to poor joint properties. The elements Fe and Al have limited mutual solubility, which can lead to the formation of brittle intermetallic compounds that can weaken steel–Al joints unless diffusion across the joint is carefully engineered. In this work, the overarching hypothesis was that AM, with rapid cooling and steep thermal gradients, along with the ability to control composition for compositionally graded joints, can be leveraged to kinetically suppress the formation of intermetallics at the steel–Al joint interface, thus enabling stronger and tougher joints. Thermodynamic, diffusion, and stress modeling approaches were employed to design compositionally graded and stepped joints from steel to Al alloys that could be fabricated by AM, as well as to understand the resistance to cracking of those joints. Two combined AM processes/joint design approaches that could potentially enable the suppression of intermetallics were investigated.

Mission Relevance

Steel and Al are some of the most widely used structural materials and are used in automotive, aerospace, marine, energy, and a host of other sectors. Steel is strong and low-cost, and Al is lightweight and has moderate strength and excellent thermal and electrical conductivity. Therefore, many systems and assemblies could benefit from robust methods to join these two materials together to take advantage of their complimentary properties. This project developed some fundamental printing strategies for multimaterial AM structures composed of steel and Al. Deployment of this technology in industry could result in stronger steel–Al joints, substantial lightweighting, and/or improved thermal management by the replacement of steel parts with printed parts composed of steel and Al in a wide variety of sectors (e.g., aerospace, light-duty automotive, heavy-duty freight vehicles, energy generation). Furthermore, the basic principles and methods developed here may be applied to other materials systems—for example, in fusion energy to make W–steel transition joints or Fe–Cu joints for thermal management applications, as well as other dissimilar material joints for a broad impact.

Results and Accomplishments

Two combined AM processes/joint design approaches were selected that could enable the suppression of intermetallics. First, ORNL collaborated with Optomec Inc. to evaluate blown powder directed energy deposition (DED) to fabricate a compositionally graded steel–Al joint that reduced the chemical driving

force for diffusion and intermetallic formation. The second approach, in collaboration with Tennessee Technological University, used a wire-based cold metal transfer (CMT) process with low heat input to fabricate Al alloy 4043 to 316L stainless steel joints with superior mechanical properties compared with 316l–Al alloy joints produced with conventional techniques. Both approaches hold significant promise for improving the robustness of steel–Al joints.

In the early stages of this work, the causes of cracking in abrupt 316l–Al alloy joints fabricated by blown powder DED was identified as the combined effect of residual stress development, coefficient of thermal expansion mismatch, and ordered intermetallics, which form at the steel–Al interface. Key technical pathways were proposed for suppressing intermetallic formation in 316l–Al alloy joints by AM, including fabricating compositionally graded joints using blown powder DED and abrupt joints using wire-based CMT AM processes. A thermodynamic and stress modeling approach was developed and used to design a 316l–4043 Al joint with reduced driving force for diffusion and intermetallic formation, as well as reduced coefficient of thermal expansion mismatch to lower thermal stresses. By controlling blown powder DED processing parameters (laser power and speed) and the cooling rate, significant suppression of intermetallic compounds, as confirmed by atom probe tomography and hardness testing, and a crack-free joint were obtained.

This project collaborated with Tennessee Technological University to identify the low–heat input CMT process as another promising route for the fabrication of steel to Al structures by DED wire AM. This work ultimately demonstrated that by controlling heat input, the intermetallic layer thickness could be significantly reduced while still achieving a sound metallurgical bond, and superior 316l–4043 Al alloy joint properties could be obtained in comparison with properties from conventional joining processes. The CMT wire-based DED process was also shown to result in lower residual stresses as measured by neutron scattering at the Spallation Neutron Source compared with a more traditional metal inert gas welding of 316l to 4043 Al. This project successfully performed the first operando stress measurements of additively manufactured bimetallic structures using the OpeN-AM system at VULCAN at the Spallation Neutron Source. These measurements provided valuable insight into the stress evolution during the printing of steel–Al joints.

NEUTRON DATA INTERPRETATION PLATFORM ECOSYSTEM

10740: Automatic Structure Refinement Platform for Neutron Diffraction

M. McDonnell, J. Liu, P. Peterson, Y. Zhang

Project Description

The overarching goal of the Automatic Structure Refinement Platform for Neutron Diffraction project was to develop a prototype software tool suite to support the workflow for real-time and automatic powder diffraction data analysis and visualization. The software tools were deployed onto the Neutron Data Interpretation Platform (NDIP) to be shared with ORNL Neutron Science user facility beamlines. These tools provide information to the experimentalist to better understand the running experiment and manually evolve the experiment plan to improve the impact of the science being explored.

Mission Relevance

ORNL hosts world-class hardware including neutron sources with high flux and cutting-edge computational power. This resource enables the real-time data reduction, analysis, and results presentation at the fundamental hardware level. However, software is yet to be further developed to catch up with the progress with instrumentation to fully use the power of hardware and present general users with fast, easy-access, and interactive real-time data analysis environment. This project will significantly improve user experience, scientific impact of experiments, and efficiency in using neutron resources.

Results and Accomplishments

Live data reduction for Spallation Neutron Source powder diffractometers was accomplished. The automated rudimentary powder diffraction pattern analysis prototypes were created for single peak fitting and pattern matching capabilities. The rudimentary powder diffraction pattern analysis tools were integrated in the NDIP to be shared by any neutron scattering workflow created on the platform. GSAS-II Rietveld and DiffPy-CMI pair distribution function refinement tools were integrated in the NDIP to be shared by any neutron scattering workflow created on the platform.

To automate live data reduction and carrying out automated rudimentary powder diffraction pattern analysis workflows, an event monitor tool was developed to watch for new files from data reduction and kickoff workflows on NDIP automatically. This prototype was used by the NDIP team to find a way to register data in NDIP produced by experiments. A prototype graphical user interface was developed for the automated rudimentary powder diffraction pattern analysis. An automated Rietveld refinement tool was created using the open-source BBO-Rietveld software.[1] The tool uses parallel processing and the hyperparameter optimization library Optuna to do a global search using GSAS-II software fitting parameters to optimize the refinement.

10741: Analysis of Mixed Samples by Monte Carlo Ray Tracing for Neutron Scattering

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Project Description

The analysis of many neutron scattering experiments is hampered by interpreting scattering from multiple processes, especially when the neutron scatters multiple times from any number of these possible processes before it is detected. Researchers have identified the use cases of diffraction under high

[1] Quantumbeam. "BBO-Rietveld." Github. https://github.com/quantumbeam/BBO-Rietveld

pressure in lanthanide metals, neutron spectroscopy measurements of magnetic excitations in $NiPS_3$, and small-angle scattering from irradiated steels encased in Pb shielding for personnel protection as covering many of the neutron scattering measurements that can benefit from the inclusion of multiple scattering in the analysis. The goal of this project was to provide an analysis tool by wrapping the McStas[2] and MCViNE[3] Monte Carlo Ray tracing packages in a user-accessible workflow leveraging multiple high-performance computing resources.

Mission Relevance

A better understanding of the scattering processes from everything in the beam is broadly needed to discern the signal from the noise. This project targeted three analysis use cases in which understanding the sample–sample and sample–sample environment interactions are critical to extract scientifically meaningful conclusions from the data. Specifically, the pressure dependence of the incommensurate structures observed in many lanthanide metals can only be addressed via neutron scattering. Scattering can provide essential baseline data to predict the performance of these materials and serves as a proxy for their actinide counterparts. The study of activated samples is relevant to producing components for nuclear energy.

Results and Accomplishments

The data acquired for $NiPS_3$ on the SEQUOIA spectrometer at the Spallation Neutron Source clearly showed a faster decrease in intensity than was observed in the spin wave model. The experimental user for this project was concerned that this result was a resolution effect. To confirm this observation, a resolution convolved model was produced using the tools developed in this project. Specifically, the incident beamline simulation tool was used to provide the dataset.[4] This dataset was then used in a resolution simulation of the SEQUOIA spectrometer, and the resolution volumes were convolved with the model. The scattering below 10 meV was suppressed compared with the model. This suppression is a possible indication of quantum damping and was published in *Physical Review B*.[5]

One of the issues with using Monte Carlo Ray tracing in the analysis of data is the speed of exectution. This project solved the incident beam caluation speed issue by moving the code to GPUs. This project tackled the next place of speed issue by moving many of the sample calculations to GPUs, which involved working a version of MCViNE[6] that leverged the Numba[7] library to expedite the calculations. For the scale of the speed-up with a commodity GPU card, the speed increased by two orders of magnitude when $>10^9$ particles were calculated, which is the typical number in a simulation.

Small-angle scattering from precipitates is challenging because the shapes can be irregular and dense. To work with this issue, this project designed a kernel for MCViNE[8] that takes as the input the Fourier transform of an image in which the intensity scale is set by the scattering length density. With this method, any image can be used to make a scattering kernel. As an example, this project took an image of an array of randomly sized octagons and Fourier transformed it. Then, this transformed image was used as

[2] P. K. Willendrup and K. Lefmann. "McStas (ii): An Overview of Components, their Use, and Advice for User Contributions." *J. Neut. Res.* 23, 2021, 7. DOI: 10.3233/JNR-200186
[3] J. Y. Y. Lin et al. "MCViNE—An Object Oriented Monte Carlo Neutron Ray Tracing Simulation Package." *Nucl. Instr. Meth. A* 810, 2016, 86–99. DOI: 10.1016/j.nima.2015.11.118
[4] G. E. Granroth et al. *Incident Beamline Simulation for the Ei=29 meV on SEQUOIA*. Oak Ridge, Tennessee: Oak Ridge National Laboratory, 2023. DOI: 10.13139/ORNLNCCS/1922042
[5] A. Scheie et al. "Spin Wave Hamiltonian and Anomalous Scattering in $NiPS_3$." *Phys. Rev. B.* 108, 2023, 104402. DOI: 10.1103/PhysRevB.108.104402
[6] J. Y. Y. Lin et al. "MCViNE—An Object Oriented Monte Carlo Neutron Ray Tracing Simulation Package." *Nucl. Instr. Meth. A* 810, 2016, 86–99. DOI: 10.1016/j.nima.2015.11.118
[7] S. K. Lam, A. Pitrou, and S. Seibert. "Numba: A llvm-Based Python Jit Compiler." *Proceedings of the 2nd Workshop on the LLVM Compiler Infrastructure in HPC 1*, 2015. DOI: 10.1145/ 2833157.2833162
[8] J. Y. Y. Lin et al. "MCViNE—An Object Oriented Monte Carlo Neutron Ray Tracing Simulation Package." *Nucl. Instr. Meth. A* 810, 2016, 86–99. DOI: 10.1016/j.nima.2015.11.118

the scattering kernel in the GP-SANS instrument at the High Flux Isotope Reactor. Notably, the result from the simulation is a neXus file identical in format to an experimental dataset from the GP-SANS instrument. Thus, the results were reduced with the same drtsans[9] tool that was used for the analysis of experimental data. The input image must have many pixels and cover a large array of points to generate a Fourier transform relatively free of artifacts. Nevertheless, for speed purposes, the Fourier transform image was then averaged to a lower resolution—one compatible with the simulation.

10818: Advanced Neutron Data Analysis for Quantum Materials

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Project Description

The goal of this project was to develop an easy-to-use framework for the advanced analysis of inelastic neutron scattering (INS) experiments on strongly correlated quantum materials and to integrate this capability into ORNL's Neutron Data Interpretation Platform (NDIP) Ecosystem. This capability is based on ORNL's DCA++ code, and the objective was achieved through the following aims: (1) streamline DCA++ simulations by developing automated workflows to integrate the manual steps required to predict the dynamic magnetic structure factor $S(Q, \omega)$, (2) use artificial intelligence (AI)/machine learning (ML) to accelerate DCA++ simulations to enable rapid parameter scans as well as future deployment on edge devices, and (3) demonstrate the developed capability for neutron scattering experiments on itinerant quantum magnets.

Mission Relevance

This project is relevant to advancing neutron sciences and computing at scale as well as in discovering the next generation of materials relevant to DOE's mission. It integrates ORNL's signature strengths in these areas by providing a new capability for high-performance computing (HPC)-based advanced neutron data analysis for spectroscopy on quantum materials and lays the foundation for the seamless integration of advanced materials modeling into an experimental neutron spectroscopy workflow.

Results and Accomplishments

The goal of this project was the development of an easy-to-use framework for the advanced analysis of INS experiments on strongly correlated quantum materials based on ORNL's DCA++ code, as well as the integration of this capability into ORNL's NDIP Ecosystem. The main project results include the development of an automated workflow for the prediction of INS spectra, a demonstration of the complete workflow and INS prediction pipeline for simple prototypical systems, and new insight into the challenges associated with AI/ML-accelerated quantum Monte Carlo (QMC) simulations and mitigation strategies.

The developed automated workflow integrates several manual steps (two DCA++ simulations, postprocessing of DCA++ data, Maximum Entropy–based analytic continuation, and visualization) of the INS prediction pipeline. This workflow was integrated into the NDIP ecosystem for a prototype demo problem in collaboration with the NDIP team. The workflow is implemented for general materials–specific Wannier tight-binding models that are generated with density functional theory electronic structure calculations and supports calculations of 2D and 3D materials models with multiple orbitals and arbitrary crystal structures. This development establishes the necessary foundation for a seamless integration of advanced, HPC-based materials modeling into an experimental neutron scattering spectroscopy workflow.

To accelerate the numerically expensive QMC solver in the DCA++ simulations, the project team developed an extension of self-learning Monte Carlo to multiorbital problems by designing a neural

[9] W. T. Heller et al. "drtsans: The Data Reduction Toolkit for Small-Angle Neutron Scattering at Oak Ridge National Laboratory." *SoftwareX* 19, 2022, 101101. DOI: 10.1016/j.softx.2022.101101

network–based surrogate model for the prediction of Boltzmann weights as well as single-particle Green's functions for individual electronic configurations sampled by the Monte Carlo algorithm. However, the training of the ML model was found to quickly become intractable because of the high dimensional input associated with the additional correlated orbitals. A different strategy was therefore pursued, in which the full QMC calculation of the Green's functions is replaced by an ML surrogate. Specifically, the team investigated a Bayesian neural network surrogate model and found that it can provide Green's function predictions with similar accuracy as the exact QMC solver. These results provide important new information into the viability of AI/ML-based acceleration strategies needed for enabling near-real-time feedback to experiments.

Finally, the automated INS prediction workflow was demonstrated for several different materials models, including a generic single-band Hubbard model, a three-band Emory model of the cuprate high-temperature superconductors, and a Kagome lattice Hubbard model of Kagome quantum magnets. These prototypical calculations demonstrate that the developed capability can enable accurate predictions of the INS spectra for quantum magnets with strongly correlated degrees of freedom using advanced HPC-based simulations.

NONPROLIFERATION SCIENCE

10313: Advanced Optical Sensors (U)

P. Evans, T. Karnowski, J. Carter, W. Ray, S. Brown

[The results of this project have been determined to be Classified. Therefore, additional information protection and distribution restrictions have been applied.]

10392: Uranium-233 Detection Science

L. Worrall, A. Krichinsky, D. Glasgow, J. Allmond, D. Archer, N. Luciano, S. Croft, R. Venkataraman, R. Mcelroy Jr., C. Romano, M. Adams, B. Roach, S. Stewart, S. O'Brien

[The results of this project have been determined to be Controlled Unclassified Information. Therefore, additional information protection and distribution restrictions have been applied.]

10417: [Title is Controlled Unclassified Information]

N. Barber, T. Karnowski, R. Kerekes, R. Henderson, C. Stanley, S. Stewart, N. Srinivas, D. Cornett

[The results of this project have been determined to be Controlled Unclassified Information. Therefore, additional information protection and distribution restrictions have been applied.]

SELF-DRIVEN EXPERIMENTS FOR SCIENCE/INTERCONNECTED SCIENCE ECOSYSTEM

This initiative did not have any completed projects in FY 2023.

TRANSFORMATION ENERGY SCIENCE AND TECHNOLOGY

10760: Mediated Aqueous Redox Flow Batteries for Long Duration Storage

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Project Description

This project developed mediation strategies to increase the energy density of aqueous redox flow batteries (RFBs). More specifically, soluble redox couples were used to mediate reversible charge storage in insoluble, high-capacity active material particles. A mediated aqueous RFB contained soluble redox couples, which were reduced/oxidized in the cell stack and were transported to external packed bed reactors containing insoluble active materials to drive mediated reactions. A reduced mediator ($H^+ \bullet M^-$) spontaneously reacted with the anode active material during charging, and the neutral mediator (M^0) was recycled back to the cell stack. This strategy provides significant advantages over conventional RFBs by decoupling the system's energy density from the redox species' solubilities. The project involved three key elements: (1) synthesis of insoluble active materials (e.g., redox-active polymers), (2) identification of new mediation schemes based on electroanalytical characterization of active materials and soluble redox couples, and (3) design of RFB hardware to collect proof-of-concept data on new mediation schemes. The overarching goal was to develop low-cost, high-energy RFB chemistries for long-duration energy storage (LDES) applications.

Mission Relevance

LDES systems are needed to enable widespread adoption of intermittent energy sources such as solar and wind. More specifically, demand is growing for energy storage systems that operate for 10–100 h at rated power. RFBs are particularly well-suited for grid storage applications because the system's energy (tank size) and power (stack design) can be independently scaled. However, major limitations of RFBs include their high cost and low energy density. This program addresses fundamental scientific barriers associated with conventional RFBs, and the overarching strategy advances DOE's mission for grid electrification and LDES needs.

Results and Accomplishments

This program characterized a library of aqueous, soluble redox couples and active materials, and these activities resulted in identification of four thermodynamically favorable mediation schemes. Schemes 1 and 2 used different mediators for charging vs. discharging to enable high utilization of active materials with sloping voltage profiles. Schemes 3 and 4 involved active materials with nearly constant voltages during charge/discharge, and in such cases, reversible ion storage can theoretically be driven with a single mediator. The efficacy of mediation schemes was evaluated in a three-electrode RFB test bed containing a 5 cm^2 cell. Here, a Hg/Hg_2SO_4 reference electrode was added to the working electrolyte tank to measure half-cell potentials. The auxiliary electrolyte relied on water splitting (i.e., H_2/O_2 evolution during reduction/oxidation, respectively) to maintain charge balance. Carbon paper current collectors and Nafion 117 membranes were used in all RFB measurements.

Mediation scheme 1 predicted that reversible H^+ storage in a redox-active polymer—that is, poly(benzoquinonyl sulfide)—could be achieved using two soluble mediators: H–anthraquinone-2-sulfonate (AQS) and 4-hydroxy-2,2,6,6-tetramethylpiperidin-1-oxyl (4-hydroxy-TEMPO, or 4-HT). To test this hypothesis, an RFB was constructed in which the working electrolyte contained 10 mAh of each mediator (assuming 1 e^- transfer per species) and 1.25 M H_2SO_4 as a supporting electrolyte. Electrochemical results demonstrated excellent reversibility and utilization for the H–AQS mediator, but 4-HT was irreversibly reduced when polarized to approximately −0.5 V vs. Hg/Hg_2SO_4. The team suspected similar unfavorable reactions could also occur with Scheme 2 containing a

tetramethylpiperidin-1-oxyl (TEMPO) mediator, so the remainder of the program focused on electrolytes containing a single mediator whose operating voltage was well-matched to that of the insoluble active material.

Scheme 3 involved a self-mediated AQS electrolyte in which a sparingly soluble salt (~0.025 m, Na–AQS) was mediated by a highly soluble form (>1 m, H–AQS). In these measurements, the working electrolyte contained 25 mAh H–AQS and 67 mAh Na–AQS (assuming 1 e^- transfer per species), which resulted in an insoluble bed of Na–AQS powder in the electrolyte tank. During the initial reduction step, a single voltage plateau (approximately −0.5 V vs. Hg/Hg_2SO_4) was observed associated with H^+ insertion into the AQS carbonyl group. The total reduction capacity was approximately 140 mAh, which was much greater than that expected for redox processes associated with only soluble AQS species (~42 mAh). This finding indicates that the soluble forms effectively mediated H^+ insertion into the Na–AQS bed. Surprisingly, the Na–AQS bed dissolved throughout this H^+ insertion process. During oxidation, the system showed high coulombic efficiency, but all AQS species remained fully dissolved. These results suggest that the mediated reaction yielded an electrochemically driven ion exchange in which the Na–AQS was converted to a highly soluble form. This unexpected finding presents an alternative method to prepare H–AQS electrolytes (typically performed over an acidic resin column such as Amberlyst 15H) for aqueous soluble RFBs. An ORNL invention disclosure detailing this discovery was filed in March 2023.

The final reaction scheme investigated in FY 2023 involved mediation of Prussian blue (PB) with K ferrocyanide (FCN), and both species had a common $Fe^{2+/3+}$ redox center. Voltammetry measurements showed that PB had an operating potential of approximately −0.2 V vs. Hg/Hg_2SO_4 with excellent reversibility and cycling stability. Similar results were obtained in an RFB cell containing an FCN electrolyte. However, upon adding insoluble PB to the FCN electrolyte, no additional capacity was observed, suggesting the mediated reaction between PB and FCN was kinetically sluggish. Therefore, future work should focus on methods to increase the heterogeneous reaction rate (e.g., by increasing the wettability of the PB surface by blending with hydrophilic components).

In summary, this program focused on developing and testing novel mediated reaction schemes for low-cost, high-energy, aqueous RFBs. Four thermodynamically favorable mediation schemes were identified and tested. Mediated charge transfer across the electrolyte–active material interface was investigated in lab-scale RFBs. Complementary activities not highlighted in this report involved developing a library of key properties (e.g., H^+ conductivity and mediator crossover rates) for several membrane formulations. The team recently reported the highest capacity to date for a mediated P anode (up to 800 mAh/g P via reversible formation of Na_xP).

10761: Operando Neutron Total Scattering for Battery Research

J. Liu, Z. Du, G. Samolyuk, M. Everett, V. Cooper

Project Description

The overarching goal of this project was to develop a suite of new operando neutron diffraction tools for battery-related research. This knowledge was used to study the local structural evolution of emerging high–energy density battery materials during dynamical cycling, including those in anode-free Li-ion batteries, Na-ion batteries, and all solid-state batteries. Particularly, this work focused on understanding the long-debated lattice oxygen redox mechanism in the ultrahigh-capacity cathodes. Efforts have also been spent on studying the compatibility between various solid-state electrolytes and anionic redox-based cathodes. The structure information obtained from operando neutron diffraction, together with density functional theory calculations, was used to guide the exploration of new types of high–energy density cathode and solid-state electrolyte materials. This work also demonstrated the plausibility of using reversible anionic redox reactions in both anode-free Li/Na-ion batteries and all solid-state batteries.

Mission Relevance

This project addresses DOE's mission by advancing the study of clean energy storage materials.

Results and Accomplishments

This project developed the first-ever high-throughput operando neutron diffraction characterization of battery materials during electrochemical cycling. This work included the development of new operando electrochemical cells and sample environments for neutron pair distribution function measurements and the optimization of data analysis and structure modeling/refinement methodologies. The team finished the design of the incident beam collimation at the NOMAD instrument located at the Spallation Neutron Source. The incident beam collimator was successfully commissioned, and the tip design and manufacture were finished. Different beam sizes were tested during in situ experiments. This project also successfully tested the in situ neutron diffraction study of all solid-state battery cells.

This project used the aforementioned operando neutron diffraction capability to understand the lattice oxygen redox reaction mechanism in emerging high–energy density battery cathode materials. This study helped the team better understand the compatibility between cathode anionic redox and liquid and solid electrolytes. The developed operando neutron diffraction tools are now fully available to the general users, including both internal ORNL users and outside users of the NOMAD beamline.

This work used the structure insights obtained from the in situ diffraction experiment to guide the exploration of new high–energy density cathode materials using lattice oxygen redox reactions. Efforts have also been spent on exploring new solid-state electrolytes that can offer better compatibility with cathode lattice oxygen redox reactions.

10821: Melt-Infiltrated Dense Anti-Perovskite Based Solid-State Batteries

M. Dixit, B. Armstrong, C. Jafta, R. Sahore, N. Muralidharan, C. Nelson, A. Abouimraine, A. Bisht

Project Description

This interdisciplinary project aimed to develop an energy-dense solid-state battery (SSB) cell through the processing and integration of (1) composite cathodes incorporating high-voltage, Co-free cathodes ($LiNi_{0.9}M_{0.05}Al_{0.05}O_2$, where M = Mn, Fe, or other metal); (2) low-temperature, processable, Li-rich antiperovskite solid electrolyte (SE; Li_3OX with X = Cl, Br, or other halogen, and Li conductivity $=10^{-3}$ S/cm); (3) ultrathin Li metal anodes; and (4) bipolar current collectors for efficient device assembly to achieve high energy and power densities. Throughout the scope of this effort, this project also leveraged and systematically employed the unique material and electrochemical characterization techniques housed and developed in ORNL facilities—specifically, advanced in situ and operando methods (using neutron, x-ray, x-ray photoelectron spectroscopy, gas chromatography mass spectrometry, high-resolution transmission electron microscopy, and others) for guiding scientific decisions through a positive feedback loop.

Mission Relevance

Energy storage systems play a crucial role in establishing high-penetration clean energy portfolios for various applications such as grid storage, electric vehicle batteries, and manufacturing power supplies. Thus, it is imperative to explore and develop innovative energy storage systems that enable high–energy density storage while ensuring safety, reliability, and cost-effectiveness. This research project addresses key challenges in SSB technology. The insights gained from this study are pertinent to both the scientific community in the SSB field and industries in the energy storage sector.

Results and Accomplishments

This project demonstrated a moderate-temperature processing protocol for antiperovskite SE materials that achieves a high degree of local control over the microstructure in processed SE films/pellets. The material processed with this developed protocol shows superior ion transport properties as well as electrochemical cycling compared with the conventional pellet. Using the distribution of relaxation time analysis, this work also reported the presence of additional relaxation processes within the high-temperature processed pellet. This research also showcased the reversible phase stability of the processed materials using in situ x-ray diffraction studies. Finally, the surface chemistry of the developed materials was evaluated, and the presence of nitrogen species was also identified. The processing and materials highlighted in this work demonstrate tremendous promise for scalable integration into high-performance, energy-dense, practical, and reliable SSBs.

10835: Solid State Battery Cells Based on Tailored Cathode Architecture

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Project Description

This project designed and built a solid-state battery cell using the following key components: (1) computational modeling to design cathode architectures for best active material utilization and electrochemical performance; (2) advanced manufacturing approaches including field-assisted freeze tape casting; and (3) advanced characterization including electrochemical testing, microscopy, and x-ray computed tomography. The goal was to design the solid-state cell sandwich synergistically integrating all of the components. The cell uses the cathode as a support element on which the rest of the components are built. Therefore, the cathode design is central to this project. In a solid-state cell, the cathode should ideally be thick and (at the same time) dense—the two criteria that appear to be in contradiction because of ion transport limitations. This project aimed to create a two-layer cathode, with the top layer designed for low-tortuosity interface with the electrolyte and the bottom layer, being dense, increasing the cell energy. This work used team expertise in battery manufacturing, solid electrolytes, numerical modeling, and characterization, as well as targeted advancement of ORNL efforts in delivering functional solid-state batteries.

Mission Relevance

This work introduces novel concepts to battery design and manufacturing, specifically in solid-state energy storage R&D, addressing DOE's mission by enabling clean energy storage.

Results and Accomplishments

As a result of this project, the team has designed and manufactured an NMC622 ($LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$) cathode that consists of two layers—the dense layer facing the current collector (i.e., energy layer) and the structured porous layer (i.e., power layer) that is cast on top of the energy layer. The goal with this work was to develop an advanced cathode for Li-ion batteries while minimizing the disruption to the manufacturing line. The structure of the cathode (i.e., distribution of the material between the two layers) was supported by numerical simulations. The simulations modeled the Li-ion battery cell containing the double-layer cathode with a polymer electrolyte [polyethylene + Li bis(trifluoromethanesulfonyl)imide, or LiTFSI] and monitored the concentration of Li in the cathode. The simulations were performed over the range of material distributions between the two layers, and the results were compared with the traditional, homogenous cathodes. In all cases, the total active material loading was kept constant at 21.7 mg/cm^2. Under 1C discharge current density, the best utilization of the cathode was achieved in the scenario in which 60% of cathode material loading was placed in the dense tape cast layer. This material distribution was followed in the manufactured cathodes.

In summary, this project delivered polymer electrolyte–based Li metal battery cells with structured high-energy NMC622 cathodes that showed advancements compared with state-of-the-art cells. The electrolyte and the interfacial resistance of the structured cathode decreased approximately five times compared with the respective values of the nonstructured cathode. Specific charge and discharge capacity are more than 50% higher for the structured cathode in all measured C-rates compared with the nonstructured, traditional cathode. The gravimetric energy density of the half-cells with respect to the NMC622 mass increased almost 100% at a C/10 cycling rate when structured cathodes were used. Stable cycling with Li metal has been demonstrated. The process requires only two coating passes with freeze tape casting applied during the second pass. Thus, this technology is suitable for batch processing of the electrodes with minimal changes to the production line.

10923: Advancing Solid-State Batteries—Incisive Characterization and Materials Design

M. Balasubramanian

Project Description

Solid-state batteries (SSBs) that feature a Li metal anode (anodeless or minimal excess Li configuration), advanced cathode composite, and solid electrolytes (SEs) are poised to make disruptive changes to the electromobility sector. However, daunting issues related to the materials' bulk and interfacial processes (e.g., electrochemomechanical degradation) and challenges in processing and manufacturing large-format devices must be overcome. This project evaluated key properties of SEs, helped advance directed synthesis, and thoroughly characterized the plethora of interfaces that are central to the operation of SSB devices. To this end, the project leveraged the advanced operando and ex situ characterization capabilities at national neutron and x-ray facilities and coupled them with electrochemical and in-house characterization methods. Such studies provided key insights on the physics and chemistry of interfaces and helped develop dynamic interfaces with persistent activity and stability, which is essential for the robust functioning of SSB devices. This work aimed to integrate earth-abundant, sustainable cathode chemistry with SSB technology, provide key insights on the mechanisms responsible for charge compensation, and unearth important structure–property–activity relationships in this important class of materials.

Mission Relevance

This work is tied to ORNL's mission in energy, but it is also relevant to missions in environmental and national security. The work is especially relevant to DOE's advanced battery research for transportation program because it will help enable SSB technology for electric vehicles.

Results and Accomplishments

The SE Ti-doped $Na_3Zr_2Si_2PO_{12}$ (Ti–NZSP) was demonstrated to be a great candidate for seawater battery applications. However, the failure mechanism is unclear when Na^+ ions pass through the SE during continuous plating and stripping. To study the failure mechanism of Ti–NZSP, a Na|Ti–NZSP|Na symmetric cell was cycled, and synchrotron x-ray tomography was performed slice-by-slice on the cell simultaneously. The electrochemical protocols included continuous plating and stripping until the cell failed. The evolution of porosity, pore size, and grayscale images of the Ti–NZSP pellet and changes in Na thickness were obtained from binarized images reconstructed by ImageJ. Preliminary results showed a damping trend in the porosity changes over run number, which indicated pore filling. Initial reversible changes in the thickness of the top and bottom Na metal indicated reversible Na plating/stripping followed by a significant decrease in the Na thickness at the bottom, which is likely because of dendrite growth that led to cell failure.

The Li_2OHCl was synthesized using a conventional solid-state synthesis approach. Subsequently, the powders were either pressed uniaxially at room temperature (RT) up to 5 tons (conventional protocol) or

were processed through a moderate-temperature process at a similar pressure. The latter involved introducing the powder to a heated pellet die (at temperatures of approximately 250°C to 300°C) and allowing the die to cool down to ambient conditions under pressure. Ex situ tomography measurements on the conventional and the processed pellets show a stark difference in the microstructures. The tomography was carried out at the 2-BM beamline of the Advanced Photon Source with a resolution of approximately 1.5 μm. The conventional pellets show approximately 12% porosity when evaluated over the entire field of view, with pore sizes ranging from several to hundreds of micrometers. However, no discernible features were visible in the pellet processed with this developed protocol. This result suggests that the high-temperature processed pellet shows a very high density (approximately 100%, as estimated from the tomography data). Achieving this level of density in a single-step process is a key advantage over conventionally processed materials either through uniaxial pressing or sintering.

Typical operando cells used for synchrotron experiments for SSBs cannot generate large stack pressure and higher temperatures, thus resulting in considerably lower performance metrics. To accurately investigate electrode–electrolyte interfaces within these systems at experimentally relevant conditions, this work demonstrated the use of a soft, electrochemically inactive material as an internal reference in the newly developed in situ cell to accurately measure the temperature and pressure at the sample location. This work used symmetric cell halide SEs to carry out the proof-of-concept studies. To the researchers' knowledge, this study is among the first attempts to develop and showcase an operando electrochemical cell for SSB research with internal measurement of temperature and pressure. The design and validation of this cell is expected to be significant to the community with the increasing research in synchrotron studies of SSB materials. The salient ideas tested with this cell design will be transferable to SSB studies using other x-ray modalities, such as spectroscopy and imaging methods.

This work synthesized Li_2OHCl, an antiperovskite exhibiting a unique characteristic—maintaining an orthorhombic phase at RT and transitioning to cubic above 40°C, affecting conductivity. To investigate the effect of temperature and pressure on the system's behavior and to help optimize the processing conditions, VULCAN neutron diffraction studies at the Spallation Neutron Source were conducted, covering four different scenarios: (1) temperature only (RT–50°C–RT), (2) pressure only (0–150 MPa), (3) simultaneous temperature (RT–50°C–RT) and pressure (150 MPa), and (4) simultaneous temperature (RT–100°C–RT) and pressure (150 MPa–unload). High-quality in situ neutron data have been obtained, and the orthorhombic-to-cubic transition has been studied in detail for various scenarios. Rietveld refinement is the next step to extract strain, crystallite size, and lattice parameters to understand the material's structural behavior under different process conditions.

TRANSFORMATIONAL DECARBONIZATION

10762: Electrochemical Non-Aqueous Carbon Dioxide Reduction with Electron-Proton Mediators

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Project Description

The goal of this project was to develop a nonaqueous electron–proton mediated flow cell system for the scalable reduction of CO_2 to high-value products, such as ethylene or CO. Nonaqueous CO_2 reduction has not been the focus of nearly as much R&D efforts as compared with aqueous CO_2 reduction despite the key advantages of using nonaqueous solvents and electrolytes—namely, that CO_2 is much more soluble in these solvents, and the energy-wasting consumption of CO_2 by hydroxide ions can be avoided. By using soluble electron–proton mediators as opposed to an electrocatalyst, the goal of this project was to independently optimize the electrode-membrane flow cell and catalysis reactor systems. Key to this approach is a rigorous understanding of the thermochemistry of proton-coupled CO_2 reduction in nonaqueous environments, which is essential for defining key metrics such as energy efficiency that must be optimized in any scalable solution. The work was split into two thrusts: (1) investigating the selectivity of various catalyst/proton mediator pairs for downselecting redox mediators and (2) developing a prototype electrolyzer with a coupled reactor bed for inducing the interaction of both mediators with a catalyst and CO_2.

Mission Relevance

This work advances DOE's mission by addressing a significant environmental challenge the nation is facing—namely, how to remove anthropogenic CO_2 from the atmosphere and then what to do with it. The scale at which this task must be accomplished necessitates further scientific discoveries and technological developments to ensure that the least amount of energy is consumed in the process. Furthermore, the knowledge and approach described herein are directly applicable to other important energy conversion processes, such as water oxidation, N fixation, and biomass valorization.

Results and Accomplishments

The team investigated the electrochemistry of CO_2 reduction catalysis using acetonitrile as a solvent because many pKa values for potential proton mediators are known in this solvent, as are many chemical reduction potentials for redox (electron) mediators. To determine the best catalyst/proton mediator pairing that would allow for the selective reduction of CO_2 at suitable potentials, the team built an electrochemical H-cell with in-line ports to the gas chromatograph (GC) so that screening candidates could be assessed quantitatively by measuring the Faradaic efficiency of the reaction. The mass flow controllers established a constant flow of CO_2 to the cathodic compartment of the H-cell that is then fed into a GC equipped with a molesieve column for the separation and quantification of H_2, CO, and CH_4 gases.

This project initially tested both Cu and Ag metal foils, but after many irreproducible experiments with Cu foils, the team focused on Ag foil as the catalyst of choice. After optimizing the cell design to control for gas leaks and reduce chemical crossover, researchers screened potential proton mediators with the Ag foil catalyst. First, cyclic voltammograms (CVs) were acquired to get a quick picture of the electrochemical response, followed by a series of controlled potentiometry experiments that allowed for the steady-state measurement of current and gaseous products. The CVs show the catalytic response of Ag foil with a triethylammonium/triethylamine nonaqueous buffer solution under He and CO_2 atmospheres. The fact that the traces overlap suggested that the introduction of CO_2 to the media did not affect the reaction; therefore, it was not surprising to find that only H_2 gas was produced during catalysis. The team hypothesized that the acidity of triethylammonium was too high for CO_2 activation to compete with H_2 production, so a less-acidic, nonaqueous proton source was chosen next in the form of $HDBU^+$

(DBU = 1,8-diazabicyclo[5.4.0]undec-7-ene). For the CV with the $HDBU^+$ buffer in He and CO_2 atmospheres, the clear shift of the catalytic current to more positive potential with the introduction of CO_2 was an indication that CO_2 was involved in the reaction, and indeed, the GC traces for controlled potentiometry experiments at all currents showed that the only gas produced was CO. The Faradaic efficiency of the reaction reached a maximum of 80% at 60 mA. This binary shift in selectivity from the more-acidic proton mediator is notable because in aqueous media, it is much more common to see a gradual shift in selectivity as the pH is changed or when the voltage is swept to more-reducing potentials. The fact that the evolution of H_2 was completely suppressed here shows the importance of understanding the nature of the interactions between CO_2, the proton donor, and the catalyst surface in this nonaqueous environment.

To that end, Tafel analysis and concentration dependence of the catalysis reaction with $HDBU^+$ were further investigated. The positive shift in the Tafel traces with increased buffer concentration is consistent with the participation of the buffer in the pre-equilibrium step(s), and the 29 mV/dec Tafel slope suggests that more than one pre-equilibrium step may be in effect. Importantly, the large change in Tafel slope between the two different proton buffers is consistent with a change in the mechanism at the surface. Further work is necessary to understand how to manipulate the chemistry at the surface to improve the rate of the reaction at lower overpotentials.

The team worked to scale the reactor design and collected data to determine the how the proton and electron mediators interact at the electrode as well as in the catalyst bed. With the data from the H-cell electrochemistry experiments, the team was able to select a Co-based reductant that had a sufficiently reducing potential to convert CO_2 to CO at a Ag catalyst in the presence of $HDBU^+$. Running the electrolysis cell at high currents (>30 mA/cm^2) with these mediators did not result in the production of CO in the reaction. Instead, H_2 was consistently observed, likely coming from reactions occurring at the electrolyzer electrode rather than in the catalyst bed. These observations point to chemical compatibility issues that will still need to be addressed to scale this technology. Further work should be focused on developing electrolyte additives to further decrease the voltage needed to activate CO_2, which should increase the window of chemical compatibilities for future mediators.

10781: Molten Salt Carbon Dioxide Capture and Conversion to Carbon Nanomaterials

S. Mahurin, B.P. Thapaliya, A. Ivanov, X.-G. Sun, T. Aytug, M. Lamm, H. Luo, G. Cong

Project Description

The development of new CO_2 capture and conversion technologies is needed to mitigate the large volume of CO_2 emitted into the atmosphere. This work proposed to develop new molten salt systems that can capture and electrochemically convert CO_2 directly into valuable products such as graphite, graphene, C nanotubes, and C nanofibers with high efficiency. To reach this goal, this project focused on the following tasks: (1) optimizing the structural, thermodynamic, and kinetic factors leading to molten salt systems with high CO_2 absorption rate and capacity and (2) understanding and controlling CO_2 transfer mechanisms at the molten salt–electrode interfaces for precise electrochemical CO_2 conversion.

Mission Relevance

This work resulted in a new capability for the efficient and sustainable capture and conversion of CO_2 to valuable products such as graphite. Though ORNL has existing capabilities in molten salt chemistry, characterization, and handling, this work extends the lab's capabilities in molten salt chemistry to the area of CO_2 separations and conversion, which is an area of global importance. This work addressed the DOE mission and the ORNL mission by developing solutions to capture and convert CO_2 to various C products useful for energy storage applications.

Results and Accomplishments

The goal of this project was to understand the mechanisms of CO_2 conversion that lead to the selective transformation of CO_2 to value-added products with a particular focus on graphite. The team constructed the experimental setup to convert CO_2 to graphite at various temperatures using pure CO_2 and different gas mixtures (e.g., CO_2/N_2) where the temperature, gas flow rate, electrode composition, and electrochemical parameters could be precisely controlled. By varying the temperature and electrochemical parameters, the team was able to produce graphite at 780°C, which is slightly lower than most literature experiments, and this graphite material showed improved energy storage capacity under fast charging compared with commercial graphite. This project used a number of characterization experiments, including x-ray scattering and gas adsorption, to show that this graphite is slightly more disordered with larger surface area compared with commercial graphite, which was responsible for the improved energy storage performance. This work was described in a publication in the journal *Carbon*.[1]

Because one of the goals of this project was to lower the temperature of the molten salt CO_2 conversion process, the team switched to a mixture of Li_2CO_3, K_2CO_3, Na_2CO_3, which had a much lower melting point than the pure Li_2CO_3 salt. The electrode composition can have a profound effect on the final C product, so this work used a variety of materials for the anode and cathode, including glassy C, Ni plate, Ni foam, stainless steel, Pt, and Cu. A Ni cathode produced optimum results for this process. One important finding of this work has been elucidating the importance of the electrode architecture in determining the specific C product. The interface between the molten salt and the cathode, particularly for the Ni, plays a crucial role in the formation of graphite because the Ni seems to play a catalytic role in the formation of graphene at the surface. Once the C layer at the surface reaches a certain thickness, the active sites of the Ni are blocked, leading to the formation of amorphous C. As a result, a higher–surface area Ni electrode results in improved graphite yield. Using the ternary salt and specific electrochemical parameters, this work was able to produce graphite at a lower temperature (~500°C) compared with other molten salt conversion processes, which was one of the targets of this project. Results are described in a publication in *ACS Applied Materials and Interfaces*.[2]

In addition to graphite, amorphous C can also be important for various applications, including energy storage, where it can serve as an electrode, and separations (liquid and gas), where it can be used as a sorbent and separations material. Using the same ternary carbonate salt, this project was able to convert CO_2 to porous C (amorphous) at a temperature as low as 450°C. An interesting result was that by increasing the temperature by a relatively small amount of 50°C to 500°C, the porosity could be modified where the pore size decreased from the mesopore range (1–13 nm) to the micropore range (1 nm). The amorphous C showed good performance as an electrode for both Li-ion and Na-ion batteries.

This project found that lowering the concentration of the CO_2 in the gas stream led to a higher gas conversion efficiency mainly because of better matching of the CO_2 with kinetic effects in the molten salt. This work achieved the objectives of the project by obtaining a better understanding of the effects of process parameters to optimize CO_2 conversion to specific targets, noting the importance of the electrode composition and interface on graphite synthesis, as well as lowering the temperature of the conversion process to graphite.

[1] B. P. Thapaliya et al. "Molten Salt Electrochemical Upcycling of CO_2 to Graphite for High Performance Battery Anodes." *Carbon* 212, 2023, 118151. DOI: 10.1016/j.carbon.2023.118151
[2] B. P. Thapaliya et al. "Low-Temperature Molten Salt Electrochemical CO_2 Upcycling for Advanced Energy Materials." *ACS Appl. Mater. Interfaces* 16, 2024, 2251–2262. DOI: 10.1021/acsami.3c14858

10812: Carbon Capture and Mineralization for Concrete Alternatives

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Project Description

The cement industry is among the world's highest CO_2 emitters despite curbing efforts by the industry (greenhouse gas emissions for Portland blended and masonry cements decreased yearly by ~15% percent since 2016[3]). To reach a net-zero CO_2 emission, a more effective approach was proposed in this project. The traditional calcination for the decarbonation of limestone is highly energy-intensive (fuel supply ~350 kg of CO_2 per ton of cement) and releases nearly an additional 500 kg of CO_2 per ton of cement in the atmosphere. Instead of a partial replacement of ordinary Portland cement by carbon-bearing supplementary material for <15%, this project aimed for near-to-100% ordinary Portland cement replacement by using carbonated cementitious materials (CCMs; i.e., a mix of carbon hydroxides, low-content polymers, and supplementary carbon-bearing waste). A net gain of ~900 kg of CO_2 per ton of cement is expected from these combined technologies enabled by additive manufacturing to inject CO_2 into the casted slurry to promote mineralization and carbon capture. The overarching goal of this project was to develop a castable CCM mix design and demonstrate its rapid stiffening for manufacturing precast elements for building construction (i.e., the concrete with enhanced durability performance and CO_2 capture efficiency).

This work combined both materials development and full-scale demonstration. The first objective was to improve the development and characterization of polymer-enhanced, carbonated, lime-based materials as alternatives to traditional Portland cement–based concrete and as a means to effectively capture CO_2 in durable construction elements. The second objective was to demonstrate a thin-walled, at-scale precast component made from CCMs that is a suitable and performant option for the exterior walls of buildings.

Mission Relevance

This project focused on the development of an integrated approach addressing both the issues of energy intensity and CO_2 emission during the decarbonation of lime and its recapture into CCM-based precast components. This research activity provided two benefits: (1) a pathway toward commercialization of additively manufactured CCM-based precast construction that enables potential future funding possibilities from DOE BTO and NETL and (2) promotes the use of CCMs for advanced additive manufacturing (full-scale 3D printed structures), a field highly captivated by the FEMP and DOE AMMTO. With a successful outcome, this technology can also potentially provide high value for the future development and rapid deployment of advanced nuclear reactors because Ca-based materials are less susceptible to irradiation-induced damage.

Results and Accomplishments

The formulations that were tested were pure hydrated lime plus polymer solutions (polyvinyl alcohol [PVA], polyethylene imine [PEI], or polystyrene sulfonate [PSS]): 4.5% PVA, 4.5% PEI, 2% PEI, 2% PSS, 2% PSS, 2% (PVA + PEI), and 2% (PVA + PSS). The 2% PVA formulation was 2% PVA per mass of dry $Ca(OH)_2$, and the solution was prepared accordingly. The slurry was cast in small beams, placed in the carbonation chamber, and removed at the specified ages: 5 h, 8 h, 1 day, and 1 week. All samples were cured in a carbonation chamber at 30°C, 40% relative humidity, and 20% CO_2 for different curing ages. The beams were then sliced to obtain a cross section for Raman spectroscopy analysis and nano indentation, and the rest was powdered for thermogravimetric and x-ray diffraction analyses.

[3] "PCA issues Industry-Wide EPD for ASTM C150, C595, C91 powders." Concrete Products. Published March 16, 2021. Accessed February 22, 2024. http://concreteproducts.com/index.php/2021/03/16/pca-issues-industry-wide-epd-for-astm-c150-c595-c91-powders/.

A separate study was run in parallel with different sample dimensions. During the initial 21-day period, an assessment of mechanical properties, including shrinkage, density, and elastic modulus, was carried out for distinct lime samples. The determination of the samples' elastic modulus was executed through ultrasonic testing. These calculations were based on the acquired longitudinal and shear wave velocities in conjunction with the material density.

The Raman map of a cross section of one of the beams (4.5% PVA) shows that $CaCO_3$ has formed at a depth of approximately half of the specimen (CO_2 exposure was from the top down for 1 week). However, the sample still showed large amounts of unreacted $Ca(OH)_2$, revealing that carbonation was taking place at a very slow pace. A summary plot of the total carbonate content formed in each formulation that was tested as a function of time shows that for the first 8 h, the carbonation rate did not change significantly. This lack of change occurred because the high content of water, which impeded CO_2 to mineralize because it first needed to dissolve in the free water in the system. After 1 day of carbonation curing, the carbonation rate started to increase as the water evaporated. That result was the case for all formulations except the sample that contained PVA. Because PVA contains an alcohol group in each chain, the samples tended to retain water, and carbonation occurred at an extremely low pace. For the other two polymers, the carbonation rate increased significantly after 1 day of carbonation curing. However, the polymers other than PVA exhibited significant shrinkage and/or cracking, and therefore, it was decided to combine two polymers to test their complementary assets. The combination of PVA and PSS showed a promising result after 1 week of exposure.

This work also examined the dynamic elastic modulus of mortars with two different sands at different ages. The slurry that was selected based on results was the slurry containing 2% PSS. The elastic modulus increased rapidly in the first 7 days, and then it slowed down. Theoretically, the material should be gaining stiffness (or strength) as it carbonates, which supports the results of this work.

Additionally, this project successfully completed the design of a CO_2 injecting mixer. Basically, a cap was designed for a Hobart mixer. The cap allows the gas to slowly penetrate the bowl while mixing the material.

The team was able to deduce that not all thermoplastic polymers are suitable for the purpose of this project and that it could be beneficial to incorporate more than one polymer in the mixture to complement the desired features. More research is needed to optimize the mortar in terms of strength and carbonation rate. Understanding the kinetics of the chemistry and the process is imperative to optimize the mortars.

10813: Adsorbent Carbon Electrothermal System for Carbon Dioxide Capture

F. Vautard, Z. Yang, H. Luo, N. Gallego, C. Contescu

Project Description

The efficiency and scalability of postcombustion CO_2 controlled capture and release using advanced carbon-based materials and electrical swing adsorption (ESA) was evaluated in this project. A carbon fiber monolith, whose surface is activated to increase adsorption capacity by generating microporosity and specific surface area, was used as the sorbent. ESA relies on the electrical resistivity of the sorbent (Joule effect) to heat it until CO_2 is desorbed. The main outcome of this project was the setup of a small-scale ESA test cell that generates data regarding the kinetics of adsorption and desorption, pressure drop, CO_2 concentration, and the electrical energy needed for regeneration.

Mission Relevance

This research assists in the development and adoption of energy-efficient CO_2 capture solutions. During the postcombustion CO_2 capture process, the regeneration of the absorbent is the energy-intensive step. This step is typically accomplished through the application of heat (temperature swing adsorption) or the

reduction of the pressure to vacuum levels (pressure swing adsorption). The intent of the project is to demonstrate that ESA is a more energy-efficient process that should help to develop industrial-scale postcombustion CO_2 capture. Potential industrial partners want to have access to a robust techno-economic analysis of the process before investing in the construction of new, at-scale CO_2 capture capacity.

Results and Accomplishments

The ESA test cell has been successfully completed and is designed for a postcombustion CO_2 capture scenario (with a concentration of CO_2 around 5%). This test cell can be adapted to other projects involving the controlled capture and release of other gases (volatile organic compounds) and/or the evaluation of alternative adsorbent designs. A full characterization of the heating process (influence of electrical power, time, gas flow) and of the CO_2 desorption process (temperature, time) was carried out. These data were used to produce a simple techno-economic model of the ESA process.

10837: [Title is Controlled Unclassified Information]

G. Veith, A. Naskar, R. Sacci, K. Nawaz, X. Zhou

[The results of this project have been determined to be Controlled Unclassified Information. Therefore, additional information protection and distribution restrictions have been applied.]

10842: Synergetic Plasma–Electrocatalytic Conversion of Carbon Dioxide and Nitrogen Gas to Urea

I. N. Ivanov, D. Hensley, Z. Wu, J. Huang, J. Jakowski, M. Li, J. Liu, V. Fung

Project Description

This project carried out synergetic plasma–electrocatalytic conversion of CO_2 and N_2 to urea, which combines advantages of electrocatalysis and atmospheric plasma technology for generation of the high flux of active intermediates. This work aimed to circumvent the high efficiency of low-temperature electrocatalytic processes by directing the high flux of active intermediates (CO and N) generated in atmospheric microplasma at the interface of perovskite oxyhydride–based catalysts (which showed superior performance in CO_2 and N_2 hydrogenation) to produce urea. These efforts advanced existing capabilities of catalytic and electrocatalytic reactions to a new area of high-energy (low-temperature) plasma catalytic processes. The project built off key ORNL capabilities in machine learning, computational modeling, catalysis, neutron scattering, and in situ spectroscopy, as well as recently acquired atmospheric plasma capabilities.

Mission Relevance

The project directly addressed DOE's mission of addressing energy and environmental challenges through scientific and technological solutions by proposing the conversion of CO_2 using scalable, nontraditional plasma reaction media, optimizing process parameters, and developing fundamental insights into factors limiting process efficiency.

Results and Accomplishments

The research team successfully grew C nanospike catalysts on both metal and ceramic electrodes, enabling direct integration into glow discharge and plasma jet setups. They confirmed the effective reactive capture of N through a plasma electrocatalytic process, where N was converted to N oxides using dry air. The rate of NH_3 generation was estimated at 88 $\mu g/(mL \cdot cm_2 \cdot h)$ with Cu nanoparticle–modified C nanospikes. Furthermore, the team achieved the coupled conversion of N and CO_2 into urea, ethanol, and alkyl carbamide using both $Cu/BaTiO_3$–xHx perovskite oxyhydride and Cu nanoparticle–decorated C nanospikes. To enhance CO_2 to CO conversion, a gliding arc plasma reactor was designed, manufactured, and tested, yielding nearly double the CO production. The reactor reached a vibration temperature of

5,000 K for N and maintained electrode temperatures around 100°C without additional cooling, though temperatures soared to 600°C for pure CO_2, indicating the need for further cooling in subsequent experiments.

10880: Intensified Carbon Capture Using Buildings Infrastructure

K. Nawaz, K. Li, R. Custelcean, B. Fricke, C. Tsouris, A. Elliot, T. Aytug, C. Finney, C. Yang, J. Brechtl, K. An, H. Jiang, D. Stamberga

Project Description

This project developed a unique concept in which the functionality of cooling towers used in commercial buildings will be expanded to provide not only cooling but also direct air capture (DAC). The overall goal of the project was to design, develop, and demonstrate a technology that can reduce the carbon capture and storage cost to less than 50% of the state-of-the-art DAC technologies. This project effort encompassed several innovations, including developing a unique contactor with unprecedented performance with acceptable fan power requirements, deploying unique fluid chemistry to enable simultaneous cooling as well as carbon capture from the air, developing a unique framework to leverage the waste heat from buildings to regenerate the capture material, and examining advanced system dynamics to ensure a sustainable operation. These innovations were achieved by using advanced multiphysics computational models, additive manufacturing of a heat/mass exchanger with optimized cooling and heating channels to manage the heat of absorption and regeneration processes, and durable and scalable surface treatments to ensure maximum interaction with the air and continual operation while processing fluid with corrosive characteristics. This project also conducted a comprehensive techno-economic analysis to ensure that the resulting infrastructure meets the cost targets. This work aimed to demonstrate a framework that can capture CO_2 at a cost that is at least 50% less compared with the state-of-the-art standalone DAC infrastructure. Through this development, the team minimized the water loss through carryover and evaporation by at least 30% compared with the existing cooling tower technology, which requires at least 1% replenishment of water per hour.

More than 10 million commercial buildings are used across the nation. Most of these buildings require extensive infrastructure for various heating and cooling processes. Cooling towers are commonly deployed in such applications in which direct and indirect evaporative cooling approaches are deployed (>250,000 installations across the nation). The goal of this work was to make these cooling towers multifunctional where they are effectively used for thermal management as well as for DAC. Notably, the distributed nature of DAC installations provides a unique opportunity to capture CO_2. A recent study by NAS has demonstrated that extensive resources are required for dedicated DAC technology, which makes such a framework highly cost-intensive, and often, the associated power requirements render DAC infeasible because the low CO_2 concentration in air requires extensive fan power to move the air through the contactors. This approach was unprecedented because it overcame two major obstacles for the widespread implementation of DAC technology (i.e., the capital cost and the operational cost). The project had many innovations that can not only realize a cost-effective DAC technology but also can improve the performance of the cooling towers for their primary function of rejecting heat from buildings. These innovations include developing advanced contactor design using topology optimization and additive manufacturing of polymer and ceramic composites to maximize the thermal–hydraulic performance, deploying surface treatment to maximize the contact between the air and the absorbent to minimize the water loss through carryover and evaporation, developing a regenerative framework leveraging the building waste heat, and designing a heat exchanger with at least a 50% reduction in size compared with existing brazed plate heat exchangers. This project deployed advanced characterization tools such as neutron imaging to understand the flow behavior in heat and mass exchangers. Another innovative aspect of this concept was system controls development, which can optimize the performance of both functions (i.e., cooling and carbon capture).

Mission Relevance

Deployment of DAC in the building sector can significantly use existing infrastructure and resources to accelerate decarbonation. Success in this project could potentially promote the deployment of this technology in numerous residential and commercial building units. The scalable, multifunctional cooling tower platform can provide a unique solution, addressing the challenges associated with higher operational cost and footprints as well as excessive heat required for the regeneration process.

Results and Accomplishments

The air contactor was systematically evaluated to understand its effect on carbon capture performance. A modular absorber device was employed to assess the efficiency of packing materials in capturing CO_2 from ambient air. This work investigated novel packing geometries for CO_2 capture from ambient air by employing 3D printed structures known as triply periodic minimal surfaces with a gyroid pattern. A comparative analysis was conducted between the gyroid-structured packing and conventional packings composed of polypropylene balls of different sizes.[4] The gyroid packings demonstrated comparable or enhanced maximum fluid capacity and pressure drops compared with traditional packings, indicating their potential suitability for industrial applications. Notably, the specific surface area (SSA) of the 3D printed gyroid structure, intentionally set at 579 m^2/m^3, was specifically compared with the smallest polypropylene packing (3/8 in.) with an SSA of 578 m^2/m^3. A notable distinction between gyroid packing and the packing with the smallest polypropylene (3/8 in.) is the significantly higher voidage observed in the gyroid structure. This disparity arises from the gyroid's flexible design, which allows for adjustments of various functional parameters such as unit cell size, wall thickness, and curvature. The higher voidage offers increased open space within the packing material, potentially enhancing gas–liquid contact and improving mass transfer efficiency during CO_2 capture processes. This information highlights the subtle differences in SSAs between the two packing types, emphasizing the potential advantages offered by the gyroid structure in terms of the surface area-to-volume ratio and its potential effect on CO_2 capture efficiency. These results demonstrated that the gyroid packing exhibited comparable CO_2 capture performance with conventional packed bed packing while significantly reducing pressure drop. This finding highlights the potential of the gyroid design as an efficient and cost-effective solution for gas–liquid contactors in DAC, contributing to the advancement of scalable and sustainable carbon capture systems.

The team developed a process model, identified various components for sustainable operation, and built the protype cooling tower facility at ORNL. Through retrofitting the cooling tower, the team turned the cooling tower into a multifunctional carbon capture device. A comprehensive online data acquisition framework was also built to monitor the real-time performance of the cooling tower. During the experiments, CO_2 concentration, pH of the solution, and pressure drop were compared for these experiments. The CO_2 concentration at the inlet and outlet was monitored using a CO_2 sensor. The test results showed a CO_2 concertation difference of around 60–100 ppm between upstream and downstream, suggesting the cooling tower can capture CO_2 from the air. Different contactors were also evaluated, and the results suggested that the wettability, surface area, and surface morphology affect the capture performance.

The results demonstrated that through using existing building infrastructure, the cooling tower can be a versatile platform for carbon capture. This platform can help solve the challenges encountered in the deployment of DAC, such as high capital cost and operational cost.

[4] K. An et al. "Direct Air Capture with Amino Acid Solvent: Operational Optimization using a Crossflow Air-Liquid Contactor." *AIChE Journal* (in press), 2023.

10925: Direct Air Capture with Aqueous Amino Acid

R. Custelcean, C. Tsouris, G. G. Jang

Project Description

This project developed a direct air capture (DAC) technology that removes CO_2 out of the air in an energy-efficient and cost-effective manner by absorption with aqueous amino acid solvents. Toward this goal, this project aimed to determine the thermodynamic and kinetic parameters of CO_2 binding and transport for selected amino acid solvents. This project applied the generated knowledge to the selection, design, and testing of an optimal air–liquid contactor, the optimization of solvent regeneration with various energy sources and minimal chemical degradation, and process design and scale-up.

Mission Relevance

DAC is a promising negative-emissions technology that captures CO_2 from ambient air using engineered chemical processes. DAC has the potential to remove legacy emissions and restore the atmospheric composition to optimal CO_2 concentration while also compensating for the current emissions from economic sectors that are difficult to decarbonize (e.g., long-distance transportation and shipping, steel, and cement industries).

Results and Accomplishments

Focusing on the aqueous potassium sarcosinate (K-Sar) amino acid, the heat of CO_2 release and solvent regeneration were determined using differential scanning calorimetry, including the sensible heat (67 kJ/mol), heat of vaporization (824 kJ/mol), and enthalpy of the reaction (117 kJ/mol). Thus, the heat of vaporization represents 82% of the total regeneration energy, pointing to the critical requirement to develop regeneration methods that avoid refluxing and boiling off the aqueous amino acid solvent. The kinetics of CO_2 uptake from air were calculated using equations developed for other aqueous alkaline solvents such as NaOH. Using this approach, the flux of CO_2 from air into a 1 M solution of K-Sar was estimated as almost twice as high compared with the NaOH benchmark ($J_{CO2} = 54 \times 10^{-6}$ mol/[m^2·s]). The CO_2 flux experimentally measured using an in-house setup was consistent with the calculated value. In a parallel effort, a cross-flow air–liquid contactor was designed using the Solidworks software, then it was additively manufactured at the Manufacturing Demonstration Facility and tested for DAC. The advantages of the cross-flow design are that it is modular—meaning researchers can easily swap the structured packing—it can be easily scaled up, and it has a minimal pressure drop. Using this setup, a number of cross-flow contactor packings were tested, including structured packing, random packing, and a combination of the two (hybrid DAC contactor [HiDAC]).

A HiDAC consisting of commercial polyvinyl chloride structured packing, enhanced with stainless steel 410 random packing, has been developed to provide a high surface area for air–liquid contact. The contactor geometry, wettability, corrosion resistance, and pressure drop, along with its CO_2 uptake efficiency, CO_2 uptake rates, and extended loading potential using K-Sar solutions were investigated. Results showed that the HiDAC contactor has a relatively high specific surface area (885 m^2/m^3), which allows for CO_2 uptake efficiencies of up to 75% and capture rates of up to 550 g of CO_2 per day for a 0.3 m wide, 0.25 m high, 0.3 m deep contactor. The contactor also exhibits high levels of wettability and corrosion resistance with amino acid–based DAC solvents. A CO_2 uptake model was developed, and modeling results were compared with experimental data to simulate and predict the performance of the contactor in a DAC process using K-Sar solvent. The results presented indicate that the HiDAC contactor is well-suited for DAC because of its high specific surface area, resistance to corrosion, and high degree of wettability. The techno-economic analysis of a DAC process using the K-Sar solvent with the HiDAC contactor found that the cost of atmospheric CO_2 removal is competitive compared with state-of-the-art solvent-based DAC technologies.

10963: Analysis of Decarbonization Pathways and Ecosystem Interactions

P. Thornton, D. McCollum, X. Shi, D. Ricciuto, J. Field

Project Description

Natural and managed ecosystems provide an important service to humanity by removing anthropogenic emissions from the atmosphere, currently protecting the Earth system from approximately 50% of the climate warming effects resulting from fossil fuel combustion and land use change. Under scenarios of decarbonization, the effectiveness of land and ocean sinks for anthropogenic CO_2 are likely to weaken. In the case of an overshoot and recovery scenario, in which climate change driven by greenhouse gas emissions reaches levels deemed unacceptably dangerous and mitigation practices are put in place to remove CO_2 from the atmosphere and place it into permanent storage, both land and ocean natural sinks can reverse sign, releasing previously stored carbon to the atmosphere. The potential magnitude, spatial distribution, and timing of such reversals have not previously been well-quantified, and the mechanisms responsible for these changes have not been adequately explored, especially for land ecosystems. Earth system and socioeconomic modeling tools exist that can be applied to the study of this basic research problem. This project used existing simulation tools to produce a quantitative estimate of the spatial and temporal patterns of land ecosystem response to strong decarbonization (overshoot and recovery) scenarios. This study explored the mechanisms responsible for changes in sink strength and sink reversals, and the corrections to socioeconomic cost models needed to account for such changes.

Mission Relevance

Relevant to the DOE Mission, this project addressed a fundamental uncertainty in the predictive understanding of the Earth system as it responds to human-caused climate change and as it further responds to the potential for human-directed climate mitigation strategies that remove CO_2 from the atmosphere. This work allowed researchers to achieve improved predictive understanding of complex biological, Earth, and environmental systems related directly to energy and included a focus on ecosystem processes that play out at the scale of organisms and landscapes while also integrating to the global scale with interactions among land, atmosphere, and ocean system components. Relevant to the ORNL Mission, this work advanced the state of knowledge associated with the potential transition away from fossil fuels toward more sustainable energy sources and, at the same time, improving global security by mitigating the damaging effects of climate change without jeopardizing the important life-sustaining functions of land ecosystems.

Results and Accomplishments

This project explored the hypothesis that a climate mitigation scenario with strong negative CO_2 emissions—a *drawdown scenario*—would result in a decreased land carbon sink because of the land ecosystem response to falling atmospheric CO_2 concentrations. Using the DOE Earth System Model, this work confirmed that vegetation carbon begins to drop immediately when the rate of change of atmospheric CO_2 declines and continues to fall through the drawdown period. Climate change mitigation scenarios need to account for this shift in ecosystem behavior.

After a few decades of reduced vegetation growth, a net improvement occurs in overall nutrient availability in the drawdown scenario, shown in this work as an increase in the fraction of potential growth that can be sustained by the available nutrient supply. This increase prevents an even greater loss of vegetation carbon during drawdown.

The drawdown scenario has the desired effect of curtailing global temperature rise, and it has the additional effect of curtailing a global rise in precipitation. These effects offset each other in terms of their effect on plant drought stress. The overall influence of climate change is a worsened plant drought stress

condition for the drawdown scenario, compared with SSP585 (indicated by the lower values for the model parameter "BTRAN" under the drawdown scenario).

Through this work, the team determined that biophysical feedback modify land sink behavior in overshoot/drawdown scenarios, placing constraints on estimates of how much greenhouse gas emissions are still permissible while keeping global climate within defined bounds of safety for society.

10965: Direct Air Capture of Carbon Dioxide Using Mineral Looping

A. Stack, J. Weber, K. Yuan; L. M. Anovitz, S. Mahurin, E. T. Rodriguez, B. P. Thapaliya, D. Sholl

Project Description

The overarching goal of this work was to establish feasibility and optimize a MgO looping process for the purposes of direct air capture (DAC) of CO_2. This goal was achieved by working to do the following. (1) Understand the controls on the rate and extent of reaction of MgO with CO_2, particularly the roles of hydrated phases in passivating carbonation reactions and their mitigation. Multiple ways to limit the formation of passivating hydrated $MgCO_3$ layers were probed, including varying the starting material, pretreatment with water, and introduction of impurities. (2) Determine the feasibility of using an air-fired calcining process and molten salt reactor to convert CO_2 into solid-phase C. A solid-phase product for CO_2 DAC would greatly simplify disposal. (3) Establish the economics of these refinements to the process to reduce the cost of CO_2 DAC. This task was to demonstrate that the refinements to the process over its original form can be performed for an economical cost, ideally improving on this method's original estimate of \$46–\$159/ton CO_2. By focusing on these three goals, this project aimed to demonstrate whether mineral looping is a valid approach to CO_2 DAC while simultaneously developing revisions to the proposed method to lower the cost and energy intensity of the process.

Mission Relevance

This project is directly relevant to DOE's mission in that the work is fundamental science with the ultimate goal to deliver a technology that will mitigate a significant environmental challenge for the United States. Specifically, prior work has suggested that this technology may have the potential to achieve DOE's Carbon Negative Energy Earthshot to remove CO_2 from the atmosphere at gigaton levels and store it at less than \$100/t CO_2 equivalent. However, significant challenges exist in implementing this technology, not the least of which is how the reaction rate of the material with CO_2 is affected by environmental parameters and material properties (e.g., relative humidity, temperature, porosity, crystallinity).

Results and Accomplishments

The principal findings in this project determined the long-term carbonation rate of MgO that was synthesized at ORNL in 1995, 27 years prior to these observations. This work determined that the carbonated reaction layer was 1.5 μm thick and consisted of multiple carbonate and hydroxylated phases, both crystalline and amorphous. This result is significant because it is a rate more than two orders of magnitude smaller than assumed in literature, and the carbonation process represents a complex reaction network rather than a single product. Reactive transport modeling allowed the team to predict that the rate slows significantly after the first year of carbonation because of long-term rates being limited by CO_2 diffusion through the reacted layer. These results are summarized in Weber et al.[5] Because of these findings, the next priority was to determine how environmental and material properties control the reaction rate. This project determined that 100% relative humidity represents a significantly faster hydroxylation of MgO surfaces on single crystals[6] and increased the rate of carbonation of MgO

[5] J. Weber et al. "Armoring of MgO by a Passivation Layer Impedes Direct Air Capture of CO_2." *Environ. Sci. Technol*. 57, 2023, 14929–14937. DOI: 10.1021/acs.est.3c04690

[6] J. N. Bracco et al. "Reaction Layer Formation on MgO in the Presence of Humidity." *ACS Appl. Mater. Int*. 16, 2024, 712–722. DOI: 10.1021/acsami.3c14823

nanoparticles at 1 atm CO_2. Furthermore, increased surface area also correlated with an increased reaction rate, suggesting that high–surface area MgO will allow a significantly faster initial rate of carbonation.

Lastly, one of the ways that the cost of CO_2 DAC can be offset is by driving the conversion of CO_2 to useful products. Thus, this project demonstrated a proof of principle to use a eutectic melt at elevated temperatures to convert CO_2 to solid-phase C. This work found that an increased conversion efficiency existed in lower CO_2 partial pressures. This result is significant because it might allow researchers to use an air-fired calciner to regenerate the MgO reactant rather than an O-fired one as previously envisioned.

10967: Modeling A Gigaton Carbon Dioxide Economy

M. Uddin, R. Clark, E. Webb, M. Langholtz, J. Thompson, M. Hilliard

Project Description

The United States needs an integrated, common vision to develop a system for capturing, transporting, storing, and using multiple gigatons of CO_2. This type of system will not necessarily evolve on its own in an efficient design, and determining which components will be the bottleneck at each stage of growth is difficult. The project created an initial version of a national-level model of the evolution of the Carbon Capture Use Sequestration and Transportation system (CCUS-T) designed for scoping and scenario development. The team proposed a strategic model that uses an agent-based simulation approach. The goal was to develop a tool that can be the foundation for a national study of CCUS-T that estimates carbon capture, transportation, use, and sequestration tonnages by location or region and by transportation mode. The team believes that the most critical decisions are not the routing and size of pipelines but the system-level decisions about the interactions between the components and the structure of incentives and markets. This model is an initial attempt to address those decisions. It is not the end point of the analysis, but it is one of the tools that could support an effort such as a Billion-Ton CO_2 Report.

Mission Relevance

ORNL is focused on research targeting critical national and global issues related to energy and the environment. The lab identified carbon capture use sequestration as a critical national technology to be developed and implemented.

Results and Accomplishments

The CO_2 supply, demand, and transport model was developed to maximize CO_2 capture while determining the potential transportation constraints that the system might encounter over the next 25 years from 2025 to 2050. The model was developed in ExtendSim and used an integrated model database. The main model database contains input and output data and contains information about the current model and entity state during the run. This approach provides the capability of an agent-based modeling platform. One of the strengths of the model is the consideration of multimodal transportation options for the captured CO_2.

The team also developed a dataset representing the point sources of CO_2, the potential transportation links and distances, a representative set of potential sequestration points, and parametric values for the simulation modeling. The simulation model matches producers and consumers based on costs, profits, volumes, and constraints. The system allows for the evolution of the market as suppliers and consumers move into and out of the market. The simulation tool allows for eventual Monte Carlo simulations to reflect the noisy nature of the market and the uncertainty of the input data.

VALIDATED DESIGN AND EVALUATION OF FUSION WALL COMPONENTS

10780: Integrated Plasma Power Exhaust Solutions for a Fusion Pilot Plant

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Project Description

This project proposed to develop the physics basis and computational capability to predict and optimize the integrated outer region of a magnetically confined fusion plasma, including the edge barrier (or *pedestal*) region and open-field line scrape-off layer and divertor region. A particular emphasis was on the interaction between highly radiative divertor scenarios required for thermal management and the edge pedestal, which must achieve a high pressure and temperature to achieve a high-performance burning plasma suitable for a fusion pilot plant (FPP). A multifidelity model for the boundary plasma was developed that, when further integrated with thermal and mechanical design of the PFCs, will enable the optimization and identification of thermal management solutions that satisfy all requirements for an FPP. These solutions included the integration of existing and emerging simplified models for each of the subsystems to allow the physics of the interactions to be studied. A new simulation capability was also developed to simulate the combined system simultaneously (guided by these physics studies) based on a new formulation of the governing equations for the boundary plasma.

Mission Relevance

The integration of power exhaust solutions with fusion-grade plasmas has been repeatedly identified as one of the most pressing challenges that must be resolved before realizing an FPP. Aside from the practical need to understand this system, the physics of both the pedestal and the divertor involve a rich, coupled, self-organized state, the exploration of which has proven quite scientifically productive.

Results and Accomplishments

A new modeling tool enabling self-consistent simulation of the coupled pedestal–scrape-off layer–divertor regions of tokamak plasmas has been developed by combining the state-of-the-art models EPED and SOLPS-ITER. A range of efficient coupling methods has been developed to improve the consistency of the profiles among the regions, including a localized surrogate model for accelerated simulations. Self-consistent calculations show promising agreement with DIII-D experiments, capturing pedestal degradation and density at the divertor detachment onset. Initial calculations in the FPP regime have discovered promising regimes for integrating a cool, radiative divertor with a hot, high-performance pedestal and core.

The project's efforts produced improvements to the software codebase ASGarD (Adaptive Sparse-grid Discretization), which allowed accurate and efficient modeling of models from plasma physics. These efforts include adding nonlinear effects of the models in question and improving the performance of the evaluation of operators though different algorithmic choices. The project also demonstrated that the codebase was able to provide accurate yet memory-efficient solutions to highly dimensional kinetic models in slab geometry. The new integrated modeling capability developed in this project was incorporated in an updated version of the IPS-FASTRAN framework.

10790: Material Plasma Exposure eXperiment Digital Twin

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Project Description

The Material Plasma Exposure eXperiment (MPEX) is being built at ORNL to address the challenges of plasma–material interactions (PMIs) in a safe, high–duty cycle operating fusion reactor. MPEX is a unique linear plasma facility that is expected to expose materials, including irradiated materials, to fusion reactor–relevant fluxes and fluences. Exploitation of MPEX will address the long-term survivability of materials in plasma-facing components in a fusion reactor. In this project, a digital twin was developed for MPEX. Three objectives were established: (1) develop a computational workflow coupling a plasma source and heating, plasma transport, impurity transport, and material science first principles and machine learning models that can calculate the background plasma and impurity ion flux, ion energy, and ion energy distribution function and material composition at the MPEX material target; (2) validate the computational workflow on the existing Proto-MPEX; and (3) demonstrate the computational workflow for MPEX. The project deliverable was a developed workflow that can calculate material target parameters from actuators on MPEX.

Mission Relevance

PMIs have been identified in many papers and reports as a key issue for obtaining a high-performance, high–duty cycle, and safe operating fusion reactorClick or tap here to enter text..[1] A recent US fusion energy community planning reportClick or tap here to enter text. recommended to support "the completion of the scientific infrastructure necessary for the study of plasma–materials interactions needed to create plasma-facing components for a fusion pilot plant by completing the MPEX and high–heat flux testing facilities."[2] The proposed MPEX digital twin will help to complete the scientific infrastructure of MPEX by accelerating the demonstration of MPEX reactor-relevant plasma fluences. No linear plasma or toroidal experiments currently test plasma-facing components, including irradiated materials, at reactor-relevant ion fluxes of approximately 10^{25} $m^{-2}s^{-1}$ and ion fluences of approximately 10^{31} m^{-2}.

To design a fusion pilot plant, the community planning report also recommends that "an essential component underpinning this effort is a strong theory and computation program, including the advancement of multiscale, multiphysics theory and modeling capabilities necessary to predict the complex interactions between numerous plasma, material, and engineering processes that will occur within a fusion pilot plant."[2] An MPEX digital twin advances multiscale, multiphysics models that are relevant to the complex interactions between plasmas and materials in initial MPEX PMI science experiments.

Results and Accomplishments

The goal of integrated modeling capabilities was achieved during the second year for a Proto-MPEX case and a PISCES-RF case. The application of various machine learning models to the experimental database was determined to be useful in predicting Proto-MPEX discharges. GITR and WALLDYN coupling was developed. The coupled COMSOL, SOLPS, and GITR model for helicon-only plasma was compared on Proto-MPEX. COMSOL, SOLPS, GITR, and WALLDYN coupling for helicon-only plasma was compared on Proto-MPEX. The first model of the PISCES-RF case was created. The chosen full workflow (i.e., COMSOL, SOLPS, GITR, WALLDYN) was applied to a Proto-MPEX case. This

[1] J. Rapp. "The Challenges of Plasma Material Interactions in Nuclear Fusion Devices and Potential Solutions." *Fusion Science and Technology* 72, 3, 2017, 211–221. DOI: 10.1080/15361055.2017.1333859. G. Federici et al. "Plasma-Material Interactions in Current Tokamaks and their Implications for Next Step Fusion Reactors." *Nuclear Fusion* 41, 12, 2001, 1967. DOI: 10.1088/0029-5515/41/12/218

[2] T. Carter et al. *Powering the Future: Fusion & Plasmas*. Washington, DC: DOE FESAC, 2020.

milestone was not fully completed because the WALLDYN simulations were not completed. Integrated modeling with coupled codes was successful for the coupling of COMSOL, SOLPS, and GITR.

10816: Simulating Transmutation Damage to First Wall Steels

T. S. Byun, Y. Lin, S. Aduloju, A. Bhattacharya, C. Kessel, Y. Yamamoto, P. Edmondson

Project Description

Attractive thermomechanical properties and demonstrated radiation tolerance make reduced-activation ferritic–martensitic (RAFM) steels the primary candidate material for fusion wall/breeder (FW/B) structures. The narrow operating temperature window (350°C–550°C) of RAFM steels, however, imposes a severe limitation on fusion reactor component design: the upper temperature limit is a result of the steep loss of creep strength with increasing temperature, and the more-critical lower temperature limit is caused by radiation-induced low-temperature hardening embrittlement.[3] Furthermore, RAFM steels will suffer from a high He generation rate—10–12 appm/dpa He—by the nuclear transmutations by 14 MeV neutrons. The elevated He concentration at a high temperature will increase cavity nucleation at microstructural features in quenched and tempered RAFM steels, thereby severely degrading key materials properties, including creep strength, ductility, fracture toughness, and swelling resistance. This study used an innovative approach based on an isotopic tailoring technique to evaluate the effect of deleterious transmutation gases on materials properties. In the absence of a 14 MeV neutron source, the approach can expand the role of the High Flux Isotope Reactor (HFIR) in the evaluation of fusion structural materials. Using the isotopic tailoring technique in HFIR irradiation, in which the Fe–8Cr steel alloyed with the isotope ^{58}Ni (1.4 wt %), which has a high (n, α) reaction (He production) cross section of ~110 mb, the research aimed to quantify the effect of He on the property loss of RAFM steels and to develop a computation capability that can predict the mechanical property degradation of steels. Research tasks were planned and executed to (1) quantify HFIR irradiation-induced microstructural changes in the RAFM F82H steels by using scanning electron microscopy and transmission electron microscopy, (2) evaluate He-induced degradation of the mechanical properties, and (3) develop barrier hardening and microstructure plasticity models and validate them using analytical microscopy and tensile property test data.

Mission Relevance

The experimental data can build the foundational science for understanding transmutation-driven material degradation in the FW/B structures. The experimental method combined with modeling and simulations bridges a critical science gap in predicting in-service performance of fusion structural materials. In identifying and quantifying the effects of He on FW/B RAFM steels and using models to translate this microstructural evolution to macroscopic property variations, this research addresses the knowledge gap identified in the FESAC report to predict and control phenomenon at the FW/B plasma–material interface.

Results and Accomplishments

The F82H RAFM steels (nominal compositions in wt %: Fe–8Cr–2W–0.1C–0.2V–0.04Ta) with isotope addition (1.4% Ni^{58}), along with other fusion materials, were irradiated in HFIR and characterized to obtain microstructural and mechanical property data. The isotopically tailored F82H steel (F82H–Ni^{58}) irradiated in HFIR achieved a much higher He:dpa ratio (~11 appm/dpa He) compared with the reference steel F82H–Ni^{60} (0.3 appm/dpa He), which yielded ~936 appm He in F82H–Ni^{58} but only ~26 appm in the reference steel at the final dose of ~86 dpa at ~300°C. Detailed microscopy was performed to confirm the high-rate production of transmutation He and overall radiation-induced microstructure changes. Both

[3] A. Bhattacharya, S. J. Zinkle, J. Henry, S. M. Levine, P. D. Edmondson, M. R. Gilbert, H. Tanigawa, and C. E. Kessel. "Irradiation Damage Concurrent Challenges with RAFM and ODS Steels for Fusion Reactor First-Wall/Blanket: A Review." *Journal of Physics: Energy* 4, 2022, 034003.

the tensile test data and the microscopy data were provided to the computational modeling task to use as input data for simulations and as benchmark data for validation of hardening and plasticity models. Based on the grain structure information from the microscopy task, a representative volume element microstructure was created to simulate the deformation of F82H steels by using the microstructure-based finite element method. A set of improved mechanical threshold stress constitutive equations was established for the calculation of stress–strain data for the representative volume element microstructure. The models and computational scheme were validated with the tensile property data for the irradiated F82H specimens. A systematic integration of these experimental and computational approaches has created a critical foundation for more practical applications.

VERTEX: ADVANCED MULTIPHYSICS SIMULATIONS FOR CORE APPLICATIONS

This initiative did not have any completed projects in FY 2023.

SUMMARIES OF PROJECTS SUPPORTED BY THE SEED PROGRAM

BIOSCIENCES DIVISION

11264: Novel Approaches to Rapidly Advance Precision Medicine

M. Garvin, D. Kainer

Project Description

The rapidly expanding multibillion-dollar precision medicine industry relies heavily on knowing the genetic mutations that underlie clinical conditions in need of treatment. If successful, the approach will result in more accurate diagnoses, reduced therapeutic failure, and fewer adverse reactions to therapy. However, even though many diseases are known to be highly heritable (i.e., because of inherited mutations), genetic markers that are predictive or causative of diseases have still not been found. This is known as the *missing heritability problem*, and if the sources of this cryptic heritability can be identified, it will have a significant effect on the US health care system in general, as well as the US Armed Forces, including the Million Veterans Project here at ORNL.

The focus of this project was to explore two understudied sources of missing heritability: epistasis and genomic structural variation. These genetic changes are not surveyed by standard methods such as genome-wide association studies carried out with common variants called *single nucleotide polymorphisms* (SNPs). Additionally, some of the methods are computationally expensive and require ORNL's unique infrastructure and algorithmic development, especially if they are to be deployed genome-wide on large numbers of individuals.

Mission Relevance

The research builds on experience gained from the development of explainable artificial intelligence and network modeling tools previously developed by ORNL under work funded by the DOE. These tools are also being used to study poplar genomes for bioenergy projects led by ORNL. Additionally, this work demonstrates the value of supercomputing to identify the genetic basis of complex biomedical conditions relevant to maintain warfighter readiness and health of the US military personnel and is therefore directly related to global security.

Results and Accomplishments

The team used genomic data from approximately 10,000 individuals provided by the Research Triangle Institute and researchers at Rutgers University to test for the role of epistasis in the highly heritable condition of cocaine use disorder, applying an adapted version of an algorithm called the Custom Correlation Coefficient (CCC) developed by Sharlee Climer at the University of Missouri–St. Louis.

The analysis focused on expression quantitative trait locus (eQTL) SNPs. These are SNPs that are statistically associated with changes in gene expression available from the Genotype Tissue Expression Project. This project identified four statistically significant eQTL-CCC networks in different ancestry backgrounds. To the team's knowledge, this research has not been done before. A large problem in genomic research is that most of the tools and information are generated from people of European ancestry and are not translatable to those of different ancestries. This work's use of eQTL SNPs as input can likely find genomic signals with smaller sample sizes, which are typical of studies of individuals of non-European ancestry. This project could have a significant, positive effect on genomic research in understudied populations.

For one of the significant genes (CYP2D6), this project was able to dive deeply into the signal using raw intensity data from the SNP genotyping arrays. Those results indicated that there was copy number variation that was associated with the use of cocaine. Additionally, the size of the genome that was affected by the copy number increase was much larger in the individuals that used the most cocaine. To the team's knowledge, this result has never been shown before for any abused substance.

11288: Multiscale Molecular Digital Twins for Electrochemical Polymers

A. Sedova, R. Sacci, B. Doughty, S. Roy

Project Description

Advances in the use of polymer-functionalized electrochemical materials are presently hampered by the inability to understand and control intricate networks of cooperative, charged molecular interactions along a range of time- and length scales—collective phenomena that emerge when charged polymer molecules are assembled at interfaces and interact with ions. Accurate, dynamic, atomic-level models of these complex charged systems would greatly enhance the understanding and success of these materials. Recent advances in high-performance computing (HPC) and artificial intelligence have unlocked the possibility of substantially higher-accuracy simulations of large systems, which also incorporate polarizability and charge transfer—details essential for understanding electrochemical processes. Digital twin models used in mechanical engineering use multiple sensors on a physical system to continuously inform a simulation model. In a similar way, this project proposed to develop a molecular digital twin for novel charged polymer–functionalized materials, which input data from multiple experimental measurements such as 2D infrared spectroscopy, isothermal titration calorimetry, and electrochemical impedance spectroscopy. Each measurement acted as a sensor for different molecular features to inform accurate HPC/artificial intelligence multiscale simulations of charged polymer systems to guide electrochemical polymer research applied to the focused use case of DNA.

Mission Relevance

Polymer-functionalized electrochemical materials form the basis for disruptive new technologies. Of particular relevance to DOE's mission is their use in the design of materials for batteries, carbon capture, critical materials separation, and molecular computing and data storage technologies. The connected experimental/HPC pipeline that this project aimed to develop would help to achieve unprecedented understanding and control of complex macromolecular systems. The development of advanced molecular sensors through this project could contribute to DOE BER bioimaging and biosensing objectives and would be responsive to biosecurity efforts. The ability to connect experimental tools facilities with ORNL's HPC facilities could help deliver solutions to problems in biosceinces, sustainability, security, materials, and computing.

BUILDINGS AND TRANSPORTATION SCIENCE DIVISION

10973: Electrocatalytic Ammonia Production with Simultaneous Methane Oxidation

T. Toops, D. Deka, A. Serov

Project Description

Electrocatalytic synthesis of NH_3 is a promising sustainable alternative to the Haber–Bosch process that can operate at atmospheric pressure and use renewable energy. Although low-temperature electrochemical NH_3 synthesis in aqueous media has seen significant research efforts in recent years, studies at high temperatures (400°C–600°C) are limited despite its potential to decrease kinetic overpotential and increase catalytic activity and Faradaic efficiency. This project focused on developing a high-temperature solid oxide electrolysis cell (SOEC) that produces NH_3 on the cathode from N_2 and H_2O; O^{2-} ions are formed in the process that transports through the oxide-conducting electrolyte to the anode. In an effort to further reduce greenhouse gases, CH_4 was fed to the anode to react with O^{2-} ions and form syngas (H_2 + CO). Such an exothermic oxidation reaction also provided heat to the cell and therefore lowered the electricity requirement. Feeding H_2O and N_2 to the same reactor allowed process intensification by removing the need for a separate H_2O electrolyzer for H_2 generation. The CO_2-free NH_3 produced electrochemically can be used in fertilizer production, pharmaceuticals, and power generation, whereas the syngas can be used for chemical and synthetic fuel production. Thus, this process has the potential to decarbonize several industrial sectors simultaneously: agriculture, chemical, energy, and transportation. The primary goal of the project was to develop active cathode and anode catalysts and demonstrate that this electrochemical pathway is possible.

Mission Relevance

This project directly ties into DOE's decarbonization initiatives and is relevant to DOE offices such as BETO, FECM, and HFTO.

Results and Accomplishments

The target of this project was to synthesize two separate sets of catalysts—one to perform CH_4 partial oxidation and the other to perform NH_3 production from N_2/H_2O—and then put them together in an SOEC to perform both reactions simultaneously. The CH_4 oxidation catalyst constitutes the anode, and the NH_3 production catalyst constitutes the cathode in such an SOEC. Both the anode and cathode catalysts were synthesized; the SOEC setup was built, thereby adding a new capability to ORNL's portfolio; and each electrode was analyzed independently. Specific progress is highlighted for each of these areas in this section.

This project has established a new solid oxide fuel/electrolysis cell capability at ORNL. The setup is able to test button cells under both single-chamber and double-chamber modes that help run specific anodic and cathodic reactions individually or simultaneously. These cells can be tested at current and voltage ranges of −10 to 10 A and −10 to 10 V, respectively, using a Biologic potentiostat across a wide range of operating temperatures.

The materials La Sr Co ferrite (LSCF), Ni-doped La Sr titanate (LSNT), and Ni supported over Gd-doped ceria (Ni–GDC) were synthesized and tested for CH_4 oxidation in a packed bed microreactor. LSCF showed good activity for complete oxidation of CH_4, but Ni–GDC and LSNT showed better activity for partial oxidation. Of Ni–GDC and LSNT, Ni–GDC is a better catalyst for CH_4 partial oxidation, providing syngas at a H_2:CO ratio of 4:1 and a low CO_2 selectivity (<10%) in the temperature range of 575°C–625°C. Because this range is the targeted temperature range in SOEC tests, Ni–GDC is also expected to show good electrochemical activity for CH_4 partial oxidation.

Cathode catalyst materials such as Pd-incorporated Ln Sr titanate were synthesized to be used in electrochemical NH_3 production tests. Button cells were prepared with yttria-stabilized zirconia electrolyte, an LSCF cathode, and various anodes to perform electrochemical CH_4 oxidation tests in an SOEC.

Various tests such as H_2O electrolysis, N_2/O_2 separation from air, CO_2 electrolysis, and CH_4 oxidation, were performed in the single-chamber cell to confirm proper working functionality. Because this method is an entirely new experimental setup, detailed preliminary tests were necessary to establish good cell fabrication and performance evaluation protocols.

10976: Harvested Coal Ash-Based Alkali-Activated Binder

D. Antunes da Silva

[The results of this project have been determined to be Controlled Unclassified Information. Therefore, additional information protection and distribution restrictions have been applied.]

10983: Efficient Long-Distance High-Power Wireless Energy Transmission

L. Xue, G.-J. Su, B. Ozpineci, O. Onar, V. P. Galigekere, E. Gurpinar, S. Chowdhury, M. Mohammad, E. Asa

Project Description

Wireless power transfer (WPT) technology constantly involves trade-offs between transfer distance, power, and efficiency. Near-field WPT systems are ideal for high power and efficiency, and far-field WPT prevails at long distances. This work aimed to push the boundary of near-field inductive WPT to a much longer distance by operating at a higher frequency but without including too many radiation effects. Both fundamental and physical coil designs were considered according to analytical and finite element full-wave simulations. GaN-based, high-frequency power electronics, along with an integrated coupler, were designed to improve efficiency and range. Preliminary experimental results showed 300 W output power over a distance of 2 m with a direct current (dc)-to-dc efficiency of 62%.

Mission Relevance

This project directly supports the DOE VTO mission to develop and accelerate the charging infrastructure for on-road and off-road vehicles and thus reduce greenhouse gas emissions.

Results and Accomplishments

This project developed the design methodology of a multimegahertz, high-power, inductive wireless charging system.

WPT involves the continuous challenges of transfer distance and efficiency. The performance factors of state-of-the-art WPT systems, including near- and far-field WPT cases, include the power transfer level, distance, and operating frequency. The solution in this study operates at or close to the near/far field boundary (transition zone). Near-field WPT systems are ideal for high-power (hundreds of kilowatts) operation, for which the power transfer distance is typically less than 1 m with over 90% efficiency. Near-field WPT has been researched extensively for short distances, and it normally operates at 85–150 kHz. Far-field WPT systems, or *wireless power beaming*, has been used in proof-of-concept demonstrations for longer distances ranging from 10 m to 1 km for medium power levels (< 35 kW), but their low efficiency (6%–54%) renders them impractical or less sustainable. This research focused on improving the power transfer distance and efficiency by operating the system at near-field and far-field transition zones.

The major challenge of the proposed long-distance WPT system was the low magnetic coupling between the transmitter and receiver. A single-loop Cu foil–based coupler was designed where the quality factor

peaks around 6 MHz, and a laboratory prototype was built. Both the transmitter and receiver coil are made of 2 mm thick, 6 in. wide Cu foils. The integrated capacitor was fabricated using the PCB process with RO3003 dielectric material. Measurements from the impedance analyzer agreed with the 3D finite element prediction of 130 pF. The PCB-based capacitor connects the two coil halves through screw contact and soldering. The full-bridge inverter board was built and connected with the transmitter coils by soldering. The rectifier (without gate drive components needed) was fabricated and connected with the receiver-side integrated coil. Both the inverter and rectifier were placed to be part of the coil to minimize the variation of the coil inductance and capacitance.

The experimental waveforms were tested at 300 W output power. The system operated at high frequency (6.4 MHz) within the near- and far-field transition zones, thus achieving a longer distance with higher efficiency than the state of the art. The project also developed a high-efficiency integrated coupler (coil and capacitor integration) along with a high-frequency, GaN-based dc-to-dc converter. The designed hardware was able to transfer 300 W of power over a 2 m distance at 62% efficiency.

CENTER FOR NANOPHASE MATERIALS SCIENCES

11257: Artificial Neural Networks Based on Memristor Crossbars of Lipid Bilayers

C. P. Collier, B. Srijanto, J. Katsaras

Project Description

This project developed a soft matter–based neural network capable of short- and long-term learning memories. Unlike previous neuromorphic networks, which consisted of soft matter–based synapses and solid-state complementary metal–oxide–semiconductor-based neurons, in this system, both neurons and synapses were assembled from droplet interface lipid bilayers. This approach replaces the three-terminal transistor, which is an active device with soft matter–based, nonlinear, two-terminal passive devices. This microfabricated device consists of up to 30 synapses connected to neurons that will ultimately be capable of distinguishing, for example, between epileptic and normal electroencephalographic data.

Mission Relevance

The work in this project is directly applicable to targeted DOE initiatives such as artificial intelligence, advanced functional materials, advanced energy systems and brain complexity, and energy-efficient neuromorphic computing.

Results and Accomplishments

The team successfully constructed a variety of 3 × 3 crossbar prototypes for use with droplet interface bilayers using the 3D printing capability at the Center for Nanophase Materials Sciences. These devices have been integrated with a 32-channel Flex patch-clamp amplifier from Tecella Instruments.

CHEMICAL SCIENCES DIVISION

10569: Enabling Combined Electrical and Thermal Energy Storage Technology

R. Sacci, J. Nanda, K. Nawaz

Project Description

This project used novel, low-cost, ion-conducting hydrogels with high thermal storage efficiency and the ability to conduct ions, serving as an electrolyte for supercapacitors and batteries. This work developed a multifunctional electrothermal energy storage prototype that incorporates phase change materials (PCMs) with high heat capacity and high–surface area carbon mat electrodes with high thermal conductivity. Together, the materials allow for efficient heat storage and recovery; however, each serves an additional purpose. The PCM can conduct ions, and the carbon mats can store electrical energy as a capacitor. Thus, novel assembly and design of the prototype into a dense form factor will enable a low-cost thermal storage device with access to standalone power during an electric power supply disruption.

Mission Relevance

Thermal management systems are essential in many national security areas, and are relevant to the objectives of EERE BTO and VTO, BES, and ARPA-E.

Results and Accomplishments

The major milestone of this project was demonstrating the design and use of a multifunctional electrolyte that enables battery material cycling and undergoes thermal energy storage.

Aqueous-based Na ion electrolytes can be developed that enable simultaneous thermal and electrochemical energy storage. This project found that a combination of Na alginate, $Na_2S_2O_3$, and borax stabilizes $Na_2SO_4 \cdot 10H_2O$, resulting in a uniform mixture that can be thermally cycled without irreversible phase segregation. Borax served as a nucleation agent that reduced supercooling and improved crystallization kinetics. The PCM composite electrolyte had a high fusion enthalpy (120–163 J/g) and a phase transition at 25.0°C with 11°C supercooling (compared with 30°C without the additives). The Na salt-based additives enabled rapid ion transport between the Na-ion anode and cathodes in both the melted and solid states. The PCM conductivity can be tuned to minimize the Na transport difference between the solid and liquid state, dropping the conductivity change from four orders of magnitude to only two. This project demonstrated that this PCM electrolyte has excellent thermal cycling stability (>10 cycles) that also maintained high room-temperature ionic conductivity (>10 mS/cm). The electrolyte enabled Na-ion cycling of the dual anode/cathode material, $Na_2VTi(PO_4)_3$, though the PCM electrolyte may have promoted decomposition, which shifted the targeted cell voltage from 1.1 V as defined by Faradaic reactions to more of a pseudocapacitor, with redox features between 0.2 and 0.8 V. The pseudocapacitor shows a dramatic reduction in capacity when the PCM temperature decreased below 35°C, which is 15°C–20°C greater than when the bulk conductivity decreased. This result suggests that Na-ion desolvation kinetics at the electrode interface become much more sluggish before the bulk electrolyte solidifies, possibly because of enhanced crystallization dynamics from increased interfacial charge densities. These properties make this composite electrolyte a promising material for multifunctional energy storage devices. Future work will involve introducing other chemical species to the electrolyte that can alter the transition pathway and kinetics of the ion transport and further optimizing the electrolyte–activate material interface to stabilize the Faradaic capacity over multiple electrochemical and thermal cycles.

11011: Uncover the Nano-Scale Design Principles for Direct Air Capture of Carbon Dioxide By Electro-Sorption

G. Rother, S. Mahurin, R. Sacci, S. Islam

[The results of this project have been determined to be Controlled Unclassified Information. Therefore, additional information protection and distribution restrictions have been applied.]

11012: Upcycling of Polyvinyl Chloride Using Liquid Metal Catalysts

F. Polo Garzon, Y. Li, Z. Wu

Project Description

Polyvinyl chloride (PVC) is ubiquitous in everyday life; however, it is not recycled because it degrades uncontrollably into toxic products at temperatures above 250°C. Therefore, it is of interest to controllably dechlorinate PVC at mild temperatures to generate narrowly distributed C materials as a platform to produce hydrocarbons. Given that PVC is a solid reactant, the use of traditional solid catalysts renders poor interfacial contact. Furthermore, homogeneous liquid-phase catalysts are not suitable for the temperatures required for catalytic PVC activation (<250°C). The purpose of this project was to use the less-studied Ga-based liquid metal (LM) catalysts to dechlorinate PVC. This project built a reactor setup where solid PVC (powder or flakes) was brought in contact with an LM catalyst in a batch or semibatch mode. The catalytic conversion was performed at 200°C. The gaseous H_2 and HCl easily evacuated the solid–liquid mixture, and the solid C material was easily separated from the spent LM catalyst by means of natural repulsive interactions. Removed Cl was weakly attached to the solid C product, and it was easily washed off with acetone at room temperature.

Mission Relevance

Reincorporation of discarded plastics into the economic cycle is key to protecting the environment while enjoying the versatility and stability of plastic products. Polymer upcycling promises to reduce the plastics discarded into the environment and the emissions of CO_2 and noxious gases generated during incineration of discarded plastics. PVC is ubiquitous in everyday life. For example, it is present in construction materials, household items, and medical supplies. Therefore, developing an energy-efficient process to reincorporate discarded PVC into the economic cycle would greatly contribute to achieving global chemical circularity. This project developed a novel approach to upcycle PVC into C materials and H_2 by using LM catalysts. The use of LM catalysts provides intimate contact with the polymeric chain, overcomes mass transfer limitations, prevents coke formation, and facilitates PVC upcycling at low temperatures.

Results and Accomplishments

This project developed a catalytic route in which a Ga-based LM catalyst was used to dechlorinate PVC (i.e., remove ~90% of the Cl content) at a mild temperature (200°C) to produce H_2; a negligible amount of HCl (a corrosive gas); and a solid, carbonaceous material in which Cl is sequestered. The Cl was easily removed from the carbonaceous material with an acetone wash at room temperature. The Ga LM promoted intimate contact between the PVC and the catalytic sites. This method is reusable, outperforms traditional supported metal catalysts, and successfully converts untreated PVC pipe.

11022: Passive Tomography for Electromechanically Coupled Composites

C. Bowland, S. Gupta, S. Venkatakrishnan

Project Description

The developed passive sensing system utilizes a multifunctional composite with integrated piezoelectric materials that generate electrical output in response to dynamic stress. Dynamic loading events alter the stress state within the composite, thereby causing a perturbation in their electrical response. These composite-generated electrical responses contain information about the dynamic loading events, which were leveraged for corresponding spatial stress-state estimation within the multifunctional composites. This project established a computational platform by developing a passive tomography algorithm for distributed stress-field mapping as a precursor of damage in the self-sensing composite from boundary voltage measurements. This effort contributes to the deployment of next-generation composites into real-world applications that are critical for infrastructure, transportation, and industrial engineering.

Mission Relevance

With increasing use of composites in various industries, monitoring composites during service will become a highly desired functionality for future composite deployment. One such application is in marine and wind turbine blades, which are under constant fatigue, at risk of impacts, and costly to take out of service for inspection. Therefore, this work addresses the missions of WETO and WPTO. Multifunctional composites are also valuable for gas storage in composite overwrapped pressure vessels, which is gaining research interest with the development of the hydrogen economy. Thus, integration of a sensing composite in a composite overwrapped pressure vessel falls within the mission of HFTO. Fiber-reinforced composites are also gaining interest for automobiles, which falls within the goals of VTO.

Results and Accomplishments

The initial tasks for the project involved developing the forward model portion of the passive tomography algorithm. The forward problem is a computational model that imparts a mechanical input on a multifunctional composite sheet with embedded piezoelectric particles and models a strain map throughout the composite. These strain data are coupled with the constitutive piezoelectric equations to calculate the voltage generated and generate voltage maps of the composite under different loading conditions. The voltage was then reported at each electrode around the border of the composite sheet and was set up in a through-thickness configuration. The number of electrodes was 32 so that each impact location generated 32 voltage measurements. The forward problem was computationally modeled for impacts at 36 locations producing 1,152 individual voltage signals that were representative of a pristine composite sample. The second part of the forward problem involved evaluating a damaged composite. Within the computation model, a defect was placed at the center of the composite by decreasing the elastic modulus of the damaged area by 1%. The strain maps and voltage maps were recalculated from location-specific impacts. The voltage signals for the undamaged composite and damaged composite were compared and revealed distinct voltage differences that were used in the inverse problem portion of the passive tomography algorithm.

The inverse model used only boundary electrode voltages to reconstruct stress distribution maps to locate induced damage. For this investigation, single-point and multiple-point defects were computationally integrated into the composite plate. The forward model was run on these composites to calculate boundary voltages, and the inverse model was run on the boundary voltages to produce a 3D reconstruction of the elastic modulus of the composite. The results from the inverse model were compared with the known locations of the damage to evaluate the number of iterations required to converge the models. The inverse model succeeded in locating damage from single-point and multiple-point defects. Images depicted damage that was placed in the composite plate at various locations and slices of a 3D reconstruction of the elastic modulus of the plate using the inverse model.

This work successfully developed a computational system in which electrical signals generated by an electromechanically coupled multifunctional composite can be utilized for damage localization to enable real-time, in situ spatial sensing of the composite. Forward and inverse models were successfully derived that converged properly to create this computational framework. This research project produced a rich volumetric dataset from a 2D voltage dataset and enabled the visualization of the stress-induced damage state in composites.

11254: A Novel Analytical Approach for the Characterization of Porous Materials

C. Gainaru, T. Saito, A, Sokolov, S. Shrestha

Project Description

New manufacturing technologies triggered the development of porous materials with optimized topologies. Because their macroscopic behavior (exploited in practical applications) can be tuned based on their many microscopic and mesoscopic variables, machine learning is currently considered to scrutinize their vast design space and guide the production of improved materials. The major bottleneck in the optimization of porous products is the absence of analytical methods that can provide large amounts of structural information in a short time and with reduced costs. Therefore, this project targeted the paucity of data about the void structures of porous materials to train the next generation of deep learning algorithms for materials design by using dielectric spectroscopy. The proposed method is novel and could mitigate the trade-off between the high structural resolution and the allocated time and financial resources of traditional approaches based on image analysis.

Mission Relevance

This concept could enable a novel capability for characterizing porous structures such as thermal insulation foams and powder-based manufactured objects under the focus of many DOE programs initiated by EERE's BTO and AMMTO as well as ARPA-E.

Results and Accomplishments

The rapid development of porous materials in recent years has demonstrated that their high surface area controlled by porosity can render peculiar mechanical, thermal, electrical, and acoustical properties that can be exploited in many technological fields. Currently, the ceramic and polymeric porous systems are already widely used for gas capture, catalysis, water treatment, sensors, molecular separation, food industry, energy storage, pharmaceutical industry, synthetic chemistry, petrochemical engineering, building insulation, advance manufacturing, and so on.

However, currently, no single experimental technique can provide simultaneous access to porosity and pore size distribution in the micrometer range and above, which is relevant to many industrial-scale applications. The proposed concept could overcome this issue of global relevance. Thus, the team aimed to develop an analytic toolbox based on dielectric spectroscopy to characterize materials with pores in a size range that is relevant for a large variety of technologies and that cannot be easily accessed using traditional methods (gas adsorption, Hg intrusion, and capillary flow porosimetries). Dielectric spectroscopy is widely employed for characterizing polymeric materials and relies on the ability of these materials to build up a macroscopic polarization under the influence of electric fields. The dielectric response of heterogeneous materials has also been studied for highly polar domains embedded in less polar matrices, but to the team's knowledge, this approach has never been used for estimating pore sizes in polymeric foams.

By exploiting the polarity contrast between two constituents composing a heterogenous material, the team proposed a concept for accessing structural information of open-cell foams. The porosity of the foam (containing mostly air with a permittivity of 1) can be trivially extracted from the measured permittivity

ε_{foam} knowing the dielectric permittivity of the bulk polymer ε_{pol} according to $\phi = \varepsilon_{pol}/\varepsilon_{foam}$. To access the sizes of the pores, these pores can be filled with a liquid (e.g., water) that contains a small amount of ions (from regular salts). Thus, the distance these ions are allowed to travel though the liquid when the polarity of the dielectric capacitor is changed will correspond to the effective size of the pores, *d*.

The dielectric measurements were performed using a modular system from Novocontrol Technologies based on an Alpha-A analyzer connected to a ZGS4 test interface. The investigated foam materials were cut as disks with a diameter of 18 cm and a thickness of 2 cm and were fully submerged for 10 min in a solution of 0.1 g NaCl salt in 100 g H_2O. Some gentle mechanical pressing was used for the complete removal of air bubbles. The sample was placed in an invar/sapphire cell inserted into a cryostat, thereby enabling a temperature stability of 0.2 K that was achieved using a Quatro controller from Novocontrol Technologies. Before foam investigation and for reference purposes, a measurement was performed using the dielectric cell filled with this ionic solution.

The results were obtained for different foams. Among other information such as porosity, the dimensions of the pores extracted with the present concept were compared with those available from electron microscopy, demonstrating good agreement. The acquisition time of the dielectric spectrum was approximately 5 min, and the overall experimental time for one sample was less than 1 h.

These proof-of-principle tests demonstrated that the proposed method could become a well-established analytical tool for accessing pore size distributions and porosity—at least for open-cell macroporous materials. The idea of accessing porous domain sizes by probing ionic displacements with dielectric spectroscopy was recently published.[1]

[1] A. Mansuri et al. "Characterizing Phase Separation of Amorphous Solid Dispersions Containing Imidacloprid." *Mol. Pharmaceutics* 20, 4, 2023, 2080–2093. DOI: 10.1021/acs.molpharmaceut.2c01043.

COMPUTER SCIENCE AND MATHEMATICS DIVISION

10922: Explaining Neural Network Models Through Graph-Based Machine Learning

S.-H. Lim, S. Kulkarni, J. Chae, C. Schuman

Project Description

This project presented a graph-based machine learning algorithm to explain similarities between black-box neural network models with a focus on spiking neural network models. To achieve this goal, this project represented neural network models as graphs, developed a graph kernel to map the graph-represented neural network in a vector space, and developed a graph similarity metric to compare neural network models in the vector space. Among different variants of neural network models, this project specifically considered spiking recurrent neural network models that are widely used in neuromorphic computing.

In machine learning, selecting the right model and tuning optimization are nontrivial challenges. Regarding why and how a certain choice works in these processes, the current understanding is still limited. Compared with other areas, DOE mission areas have the unique advantage of rich scientific domain knowledge and experts. Thus, integrating such domain expertise with the proposed method to understand machine learning models can offer the opportunity for advanced interpretability. The advanced interpretability will enable researchers to obtain new scientific knowledge from machine learning models and to systematically diagnose and compose machine learning models. Specifically, the graph-based approach in this project can uniquely provide (1) contextual information of identified interesting regions in a neural network model and (2) discrepancy metrics to compare substructures of models in a computationally efficient way.

Mission Relevance

In general, this project established computer science and mathematical capabilities in interpretable machine learning with the focus on graph data, which is part of priority research direction 2 (interpretable scientific machine learning) in DOE ASCR's BRN report for scientific machine learning.[1] The report highlights the importance of interpretability in high-regret, safety-critical systems with three major research thrusts: (1) interpreting the data complexity, (2) creating interpretable model introspection, and (3) describing differences between models.

Results and Accomplishments

Evaluation results for 5,000 spiking neural network models from an autonomous vehicle use case show 82% of testing accuracy to classify models as worth training or not only based upon their model descriptions before training and testing the model. Additionally, the developed visual analysis algorithms on top of the proposed methods effectively visualize similar neural networks compared with state-of-the-art techniques such as Feather graph, Wavelet characteristics, and GL2Vec. These results were published in multiple machine learning venues.[2,3,4]

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[1] N. Baker et al. *Workshop Report on Basic Research Needs for Scientific Machine Learning: Core Technologies for Artificial Intelligence* (Washington, DC: DOE SC ASCR, 2019). DOI: 10.2172/1478744.
[2] R. Patton et al. "Neuromorphic Computing for Scientific Applications." 2022 IEEE/Association for Computing Machinery Redefining Scalability for Diversely Heterogeneous Architectures Workshop, IEEE, 2022.
[3] Z. Lu et al. "Attention for Causal Relationship Discovery from Biological Neural Dynamics." Causal Representation Learning Workshop at NeurIPS 2023, 2023.
[4] G. Cong et al. "Hyperparameter Optimization and Feature Inclusion in Graph Neural Networks for Spiking Implementation." 2023 International Conference on Machine Learning and Applications, 2023. "SNNVis: Visualizing Graph Embedding of Evolutionary Optimization for Spiking Neural Networks." Planned to submit to IEEE Visualization 2024.

10995: A Model for Quantifying Resilience and Understanding Plant Responses to Stress

J. R. Osorio, J. Restrepo, D. Weston

Project Description

This work addressed the problem of quantifying ecological resilience in biological systems, particularly focusing on noisy systems responding to stress via sudden adaptations. The motivation was the dynamical response of resilient plants. The team incorporated techniques from nonequilibrium statistical mechanics to formulate a measure of ecological resilience through adaptation specifically tailored to noisy, forced systems that undergo physiological adaptation because of stressful environmental changes. The approach used stochastic linear response theory to compute how the expected success of a system, originally in statistical equilibrium, dynamically changes in response to an environmental perturbation and a subsequent adaptation. The resulting mathematical derivations enable the estimation of resilience in terms of ensemble averages of simulated or experimental data.

Mission Relevance

Resilience refers to the ability of a system to maintain a certain degree of functionality during disturbances. By understanding and measuring the resilience of ecosystems, agricultural systems, and other biological entities, scientists and policymakers can develop more effective strategies to mitigate the effects of climate change, natural disasters, and other environmental stresses. This quantification enables the identification of critical thresholds and tipping points, thereby informing decisions about resource allocation and risk management. Moreover, it aids in the development of transformative science and technology solutions that can enhance the ability of these systems to adapt and recover from disturbances. This ability is essential for maintaining food security, protecting biodiversity, and ensuring the sustainability of natural resources, all of which are vital components of a prosperous and secure nation.

Results and Accomplishments

The results are synthetized in a research paper in *Physical Review E*.[5] The team theoretically derived and illustrated a novel proposal for a measure of ecological resilience through adaptation. The relevance of the resilience measure to biological systems rests upon the assumption that the time evolution of the biological system is described by a stochastic differential equation with an initial stationary probability distribution (before the application of a stress). Unlike most existing measures of resilience, this measure suggests that the time history of a well-chosen success observable is essential to the determination of the organism's resilience to adaptation. Via a probabilistic approach, the team captured resilience as an ensemble mean among a population of individual subsystems that can have different starting conditions and random responses to stress.

Specifically, the team developed a probabilistic measure of the degree to which a system in statistical equilibrium can dynamically adapt, recover, or change equilibrium distribution after a stressful perturbation on the parameters modeling its environmental conditions. Here, the word *adaptation* is used in the physiological sense, namely as the ability of living systems to adjust their dynamics (e.g., metabolism) in response to their changing environments. The degree of recovery is conceptualized in terms of a success function of the state variables designed to model performance, health, and productivity; the modeler seeks to maintain as high a value as possible for this measure on average for any given environmental conditions. The probabilistic approach enables dynamic models of aleatoric, epistemic, or measurement uncertainty (e.g., observations and parameters in stochastic differential equations) and enables assessment of resilience of an aggregate or ensemble of subsystems exhibiting random variations on their state variables.

[5] J. M. Ramirez et al. "Probabilistic Measures for Biological Adaptation and Resilience." *Physical Review E* 109, 2024, 024413. DOI: 10.1103/PhysRevE.109024413.

The team provided an example of a very simple model assuming low-dimensional gradient flow dynamics and forced and adapted the dynamics by simple changes upon the potential. For that example, the team demonstrated that its measure of ecological resilience is compatible with existing approaches and captures well the ability of the system to adapt after a bifurcation-inducing perturbation.

Among the contributions outlined in the manuscript is a theorem with a novel representation for the expected gradient of an observable with respect to changes in the parameters of the underlying stochastic differential equation. This result represents a contribution to linear response theory as an effective tool for studying the response of nonlinear noisy dynamical models.

The team's theoretical developments are general and applicable to biological systems that change over time and that have inherent hierarchies of biological organization. For real-life biological systems, assessing resilience will involve a series of interconnected dynamics described by mathematical or data-driven models (e.g., modeling cell to leaf, leaf to plant). The team's proposal for resilience to adaptation will apply to these more complex systems if assumptions regarding the stationarity of distribution of the state vector dynamics hold. The team proposes specific methodologies to estimate resilience using actual experimental, field, or simulation data.

11013: Skin Cancer Classification System with Mobile Devices

H. Yoon, J. Hinkle, F. Alamudun

Project Description

Reflectance confocal microscopy (RCM) is a scientific modality in biomedical imaging that provides highly advanced and noninvasive techniques for acquiring high-resolution images of the skin and superficial tissues. It has become an important tool in clinical dermatology, revolutionizing scientific imaging. One of the key advantages of RCM is its capability for in vivo imaging, allowing the capture of live tissue images without the need for biopsies or tissue excision. This ability has transformed it into an indispensable tool in modern scientific diagnostics, providing less invasive and more accurate methods in dermatology. This project used artificial intelligence and machine learning algorithms to interpret RCM images. This endeavor aimed to provide dermatologists with enhanced decision-making tools.

Mission Relevance

This project involved training models on high-performance computing environments and focused on exploring and implementing new algorithms for self-supervised training. Specifically, this project used transformer-based image comprehension models to achieve image clustering and classification tasks. These advancements have the potential to significantly enhance the understanding of scientific solutions in advanced scientific computing and health data sciences.

Results and Accomplishments

This study contributed to the synergy of RCM and artificial intelligence in the evolving scientific landscape by creating a novel segmentation strategy for RCM images. This work focused on textural features to identify key clinical regions, aiming to enhance scientific accuracy and efficiency.

The algorithm consisted of the following steps.

1. Image patch generation: The RCM image was divided into several image patches with a resolution of 256 × 256 pixels. This division allowed researchers to capture fine details and maintain image features in the corners by applying overlap and flipping on boundaries.
2. Image feature extraction: In this step, a machine learning model was trained to predict numerical representations of the images, which are referred to as *image features*. To achieve this, the team used self-supervised learning with the DINO algorithm to train a Vision Transformer model. This

approach enabled researchers to extract meaningful information from the images and learn their underlying characteristics.

3. Clustering image features: To group image patches that exhibit similar image features, the team employed the k-means clustering algorithm. This clustering process helped to identify patterns and similarities within the RCM images. To ensure accuracy, expert clinicians reviewed and determined the characteristics of the image features, providing valuable insights for labeling the clusters.
4. Create feature map: Finally, the team reflected the image feature map onto the original image, allowing researchers to visualize the extracted features in their original context. This feature map provided a comprehensive representation of the image, enhancing researchers' understanding of the underlying structures and textures.

This research demonstrates the clinical relevance of the feature map and the potential use of the cluster map as a segmentation tool for RCM images. This approach could enhance the effectiveness of dermatologists in reading cases and accurately diagnosing skin conditions.

11224: Automating Data Preparation for Science with Skel Intelligent Planner

J. Logan, J. Y. Choi, M. Wolf

Project Description

The overarching goal of this project was to connect the creation, reuse, and sharing of scientific artificial intelligence (AI)/machine learning data pipelines to a modern, engaging, and ergonomic user experience of a rich computational toolkit. The Skel Intelligent Planner (SKIP) project used a model-based approach to assemble, manage, and deliver to users reusable AI data processing workflows. In particular, users find it challenging to incorporate advanced data management features such as automated data characterization and synthetic data generation into their pipelines for generating training datasets in a straightforward and reusable way. The SKIP system created in this project demonstrates an easy-to-use, reusable, user-centric way of delivering such capabilities for ORNL science.

Mission Relevance

This work helps enable explainable, reusable processing environments for AI training datasets. Leveraging this work will allow ORNL to deliver scientific discoveries and technical breakthroughs more quickly and reliably in support of ORNL's mission.

Results and Accomplishments

To drive these efforts, the team created a data test bed with datasets at multiple scales and sizes, in several different formats, and representing multiple science domains.

As a primary science driver, this project constructed several datasets consisting of images created with the Gray–Scott benchmark application. Gray–Scott is based on a parameterized model of reaction–diffusion equations and exhibits a range of linear and nonlinear behaviors, making it a useful system for creating realistic images that reflect a range of different science applications. The team used the Gray–Scott code to create image collections at different sizes, with images exhibiting different behaviors, and in color and grayscale.

Though Gray–Scott was the main science driver for these efforts, this project also leveraged a range of other image and tabular datasets to ensure that SKIP is capable of compatibility with other data types. Among these datasets, this project used images from the MNIST (Modified NIST) database that provide handwriting samples suitable for training handwriting recognition tools. This work used a variety of tabular environmental datasets stored as comma-separated values (.csv). This project also leveraged

entries from the MIMIC-III (Medical Information Mart for Intensive Care) Clinical Database stored as .csv files.

The team created the SKIP platform to serve as a home for a set of modular, plug-in–based data analysis and wrangling tools. SKIP is a multitiered environment that connects an interactive, browser-based graphical user interface with a generative, back-end execution engine capable of running on a remote system, cluster, or supercomputer. The base system has support for tabular data and image collections, but SKIP's modular design allows extensions to be plugged in, offering additional functionality or alternative implementations.

SKIP consists of two main components, which were both created during this project. SKIP's computational component (i.e., back end) is designed to run in a remote setting, which might be a workstation, cluster, or supercomputer, but the computation component generally runs on a platform with sufficient memory and computing resources. Written in Python, the back end uses the flask library to act as a server for SKIP's REST application programming interface. The server accepts a rich set of requests that allow the user to access data or associated metadata and perform various data-wrangling operations.

SKIP's other main component is the front-end client, which runs in the user's web browser and provides a friendly interface to data and models managed by SKIP. This project implemented a flexible front end using the Vue JavaScript library. The client provides a browsable catalog of active datasets and offers access to a data type–specific collection of operators that can be applied to each dataset.

One key requirement was to address the need for human-in-the-loop support for long-running computations. SKIP's design allows for a simple mechanism to support human callbacks as long as running operations can be initiated in the front end, and a result of such an operation will appear as a new dataset that can be accessed via the catalog. Further improvements are possible, including push notifications to alert users for tasks requiring rapid response, and will likely be added as needed by future data use cases. Additionally, the team envisions future plug-ins tuned to specific tasks (such as labeling) and has created the underlying structure to support such plug-ins.

The project's other main thrust was around the creation of a variational autoencoder (VAE)-based tool for data synthesis to serve as a first example of a deep capability made available as a SKIP plug-in. This data synthesis plug-in operates on image collection datasets, with individual images in a dataset being used to train a deep learning model, which may then be used to generate synthetic images that are similar to the images in the original dataset.

The VAE tools serve as a good example of the type of functionality that SKIP is designed for. First, it works directly with SKIP's native image collections and allows the use of the PyTorch-based deep learning capabilities without requiring the user to write any code. Instead, the system asks a few basic questions about the desired VAE model and uses a set of code templates to generate a set of Python codes that support creating, training, and using the VAE model. The user interacts with all functionalities through a graphical user interface. The underlying code is available for inspection but is not central to the user experience.

Finally, to show how SKIP could be used to achieve more complex data wrangling goals, this project assembled a demonstration focused on the Gray–Scott data and a useful collection of SKIP operations. The demonstration included importing several datasets and performing clustering on that data to understand what image classes were well-represented and which were rare. With this information in mind, the user could leverage the VAE tools to create a model of the images in the collection with training proceeding in one or more steps and including visual and statistical feedback on the model. Once sufficiently trained, that model was then used to create synthetic images to augment the dataset with similar but unique images.

FUSION ENERGY DIVISION

11250: Liquid Metal Blanket Candidate for the United States Fusion Pilot Plant

S. Smolentsev, P. Humrickhouse, J. Fan, S. Aduloju, C. Kessel

Project Description

Selection of the right breeder and blanket concept is vitally important to the successful development of the US fusion pilot plant (FPP). At present, PbLi is considered as the most promising liquid breeder candidate. The dual-coolant PbLi (DCLL), He-cooled PbLi, and water-cooled PbLi blankets are the world's liquid blanket concepts proposed by up to seven nations. The associated blanket designs are mostly adopted to a tokomak reactor with large, D-shaped toroidal field coils. The most critical feasibility issue of such blankets is high magnetohydrodynamic (MHD) pressure drop owing to a strong magnetic field of 5–10 T and MHD flow–induced corrosion. The anticipated new features of the FPP include an even stronger magnetic field up to 20 T, compactness, and demountable magnets. These features may add more limitations on the blanket design but can also provide more design opportunities. This project performed feasibility studies for the conventional DCLL blanket for strong magnetic fields of 10–20 T, which have not been investigated yet. Then, a self-cooled blanket concept called a toroidally symmetric PbLi (TSLL) blanket (which was also proposed in this project) was assessed. In the TSLL, mitigation of the MHD pressure drop is envisaged through a toroidally symmetric flow configuration, making use of the demountable magnets. The results of the proposed exploratory studies will help in selecting blanket candidates for the US FPP and in designing and constructing new liquid metal experimental facilities at ORNL.

Mission Relevance

The project advances the DOE Mission to ensure America's security and prosperity by addressing its energy, environmental, and nuclear challenges through transformative science and technology solutions by proposing new breeding blanket concepts and their analysis.

Results and Accomplishments

Although the FPP concept is being evolved, the common view is that a stronger magnetic field compared with present fusion devices' magnetic field B will be used for plasma confinement. All previous blanket studies have been limited to $B < 10$ T. In this project, new results of integrated studies for a DCLL blanket were obtained for $B = 10–20$ T. The blanket geometry, materials, and thermal loads were adopted from the existing DCLL design for the US Fusion Nuclear Science Facility. The computed results of MHD flows, heat transfer, and corrosion for the reference DCLL blanket at PbLi $T_{in}/T_{out} = 350°C/550°C$ suggest that the maximum allowable magnetic field is 16 T. High-temperature DCLL was found to exhibit acceptable MHD pressure drop even at B = 20 T, but the corrosion losses may exceed the conventional limit of 20 μm/year. The analyses suggest that the DCLL blanket is a prospective blanket candidate for future fusion devices with a very high magnetic field >10 T.

In this project, a new self-cooled liquid metal blanket concept called the TSLL blanket was proposed and assessed, including analysis for MHD flows, mechanical analysis, and heat transfer and neutronics assessments using the ARC (Affordable, Robust, Compact) reactor with demountable magnets designed by the Commonwealth Fusion Systems as a test bed. The proposed blanket uses a PbLi alloy as the breeder/coolant and reduced activation ferritic/martensitic steel as structural material. A special feature of the new concept is the toroidally symmetric flow in the blanket-integrated first wall and the breeding zone to reduce the MHD pressure drop, as well as the use of anchor links to strengthen the first wall construction. The provided estimates suggest an acceptable MHD pressure drop, required mechanical integrity, and high tritium breeding ratio. As a result of these assessments, the new blanket concept can be

recommended for more detailed studies as a promising blanket candidate for implementation in future fusion devices.

MANUFACTURING SCIENCE DIVISION

10946: Silicon Nitride Fiber Development

S. Bullock, S. Yilmaz

The first objective of this project was to develop candidate polymers for creating Si_3N_4 fiber. The primary candidate is cyclosilazane. Reducing the oligomer in an NH_3/N_2 gas mixture up to 350°C makes an amorphous polymer suitable for spinning. Processing in an NH_3/N_2 atmosphere at 1,400°C can create semicrystalline Si_3N_4-f. The second objective focused on the polysilsesquioxane polymers as the fiber precursor converted to Si_3N_4-f via carbothermal reduction with NH_3. Polysilsesquioxanes have been used to create SiC fiber precursors with melt spinning.

Mission Relevance

An urgent need exists for these materials and the fiber precursor for use across a variety of industries.

Results and Accomplishments

Polysilazanes of varying molecular weights were produced. The reactions that ran at 250°C made polymers that were viscous, and reactions performed above 350°C made solid polymers. To meet the milestone, 1,500 g of liquid polymer were produced over the course of the project. The Parr-type reactors are scalable to 10 L quantities and can produce 10 kg. The thermal polymerization process is scalable. The monomer-to-polymer conversion was confirmed via ^{13}C and ^{29}Si nuclear magnetic resonance. The monomer peaks at −5 ppm were not present sin the runs after heating to 350°C. This lack of a peak is verification of polymerization owing to the ring opening of the structures to a linear polymer. Melt processing of the polymer proved difficult because of the polymer's high melting point. The differential scanning calorimetry trace showed a poor melt transition up to 400°C. This poor transition indicates that the polysilazane is not easily melt-processable in a useable range of 300°C, similar to thermoplastics. This polymer is more amenable to solution spinning; the synthesized polysilazane is very soluble in toluene (25 wt %). Thermogravimetric analysis provided a respectable yield of 58% based on a 20% vinyl fraction. This thermogravimetric analysis compares favorably to prior ceramic yields from polysilazanes, which reported a 20% yield to 1,000°C.[1] Melt rheology was not achievable based on the high melting point and limits of the Ares GA rheometer. Gel permeation chromatography was not available initially because the only solvent was tetrahydrofuran, and polysilazane was not soluble in tetrahydrofuran. The polysilazane was, however, soluble in toluene. Gel permeation chromatography was performed using toluene as the mobile phase, and the molecular weight was 2,134 g/mol using a polystyrene standard as a reference. The 2,134 g/mol molecular weight was similar to other literature reports.[2]

10984: Efficient Microwave Sintering of Additively Manufactured Ceramics

G. Larsen, S. Bullock, T. Aguirre

Project Description

This work used microwave furnaces to process additively manufactured (AM) SiC ceramic parts. AM was used to make near-net-shape parts in complex geometries, which are often otherwise unable to be manufactured. Microwaves were used to quickly and volumetrically heat the parts, enabling faster and more energy-efficient processing. Lastly, this work leveraged experience from the team on cross-linkable preceramic polymers to reduce the number of polymer infiltration and pyrolysis cycles needed.

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[1] Seyferth, D., and G. Wiseman, *A Novel Polymeric Organosilazane Precursor to Si_3N_4/SiC Ceramics*, ONR CONTRACT N00014-82-K-0322, Task No. NR 631-618 (Cambridge, Massachusetts: Massachusetts Institute of Technology Department of Chemistry, 1985).
[2] Wiseman, D. S., and H. Gary, *A Novel Polymeric Organosilizane* (Washington, DC: Office of Naval Research, 1985).

Mission Relevance

Silicon carbide is broadly important because it is stable under high temperatures and harsh conditions. The material is currently used in turbines for electric power generation, as well as in aircraft engines, nuclear reactors, and heat exchangers. Cost, derived largely from processing, is a major barrier to wider adoption of SiC. This work helped to address the speed and geometries that can be formed with the intent of increased use in power generation and heat exchanger applications.

Results and Accomplishments

The work on this project showed the potential of microwave processing of AM SiC but also some unexpected challenges. The SiC parts were effectively produced via binder jet AM to make green preforms. Preceramic polymer infiltration and pyrolysis showed good ability to infiltrate parts, and the SiC is highly absorptive at the 2.45 GHz frequency. The primary challenges were maintaining an anaerobic environment for cure and pyrolysis and handing the high off-gassing rates of explosive gases from polymer infiltration and pyrolysis resins because of the high heating rates of the microwave systems. The microwave system used in this work was capable of heating parts at more than 30°C/min, which caused all the evolved gas from the preceramic polymers to be released within minutes. Additional work is needed in drying ceramic slurries and sintering alumina fiber–based ceramics.

11040: In Situ Heat Control Strategies for Fast Exothermic Catalytic Reactors: Catalytic Material Design Informed By the Numerical-Modeling

C. Karakaya, M. Kidder, Z. Mills, B. Adkins, T. Turnaoglu, T. LaClair

Project Description

This project aimed to develop model-based, in situ heat control strategies for fast, exothermic, heterogeneous catalytic reactions. The concept was demonstrated through a bifunctional material design, which was used as a catalyst and as a heat sink to control the reaction exotherm. The interconnected nature of chemistry, heat, and mass transport in a packed-bed reactor was studied through numerical modeling.

In general, for a fast, exothermic chemical reaction, the chemical timescale is much faster than the heat transfer timescale. Because most heterogeneous catalysts use ceramic supports with relatively low thermal conductivity, the net heat release is often localized, resulting in the formation of hot spots that lead to stress and unrecoverable physical damage to the catalytic material. Furthermore, the uncontrolled reaction heat leads to side reactions, which eventually decrease the product yield.

The oxidative coupling of methane (OCM) was chosen as an example catalytic process to study the nature of a highly exothermic ($\Delta H°_{298} = -281$ kJ/mol) catalytic process. The OCM process is a direct pathway to form ethylene, which is a highly valuable chemical and building block material of fuels.

When successful, the ethylene yield and production rate was expected to increase by establishing in situ temperature control and avoiding the side reactions leading to CO_x. The results derived from this study were used to develop nondimensional parameters defining the relationship between the thermal and chemical characteristics of the system. These parameters, which are independent of the specific process, can be used in the design of catalytic systems capable of maintaining isothermal operation.

The main objective of this project was to improve the performance of chemical reactors by coupling smart material design with modeling and simulations. The example studied here was OCM, but the concept is applicable to any chemical process in which the heat and mass transfer limitations hinder the process yield.

Mission Relevance

Most fuel conversion technologies (e.g., Fischer–Tropsch, C–C coupling, CO_2 hydrogenation, and aldol condensation reactions) are exothermic in nature and require precise temperature control. An uncontrolled heat of reaction leads to thermal runaways, resulting in a decrease in product selectivity and possible coke formation. Although most chemical processes (e.g., Fischer–Tropsch) rely on ex situ temperature control techniques, the interconnected nature of chemical reactions and heat transfer processes requires a fundamental understanding of the complete process at the microscale to control the rise in localized temperature and subsequently avoid the unfavorable side reactions. Because fast and highly exothermic chemical processes are common in fuel conversion, reliable modeling approaches to analyze the relationship between the chemical and heat transport timescales are required, thereby optimizing the yield through material design. OCM was chosen as a representative example of this challenging class of processes. The fundamental understanding derived from this study is applicable to other exothermic processes similar in nature that require precise temperature control.

Results and Accomplishments

The concept of core–shell design was proven to be a reliable methodology to control the reaction temperature in a highly exothermic reaction environment (e.g., OCM). Although the suggested materials are specific to the OCM process, the methodology and the numerical model can be used to study various heterogenous chemical processes.

A 2D particle model was developed in COMSOL Multiphysics software. This model was used to design the core and shell properties of the catalyst particles and to optimize the heat and mass transfer characteristics in a 2D reactor geometry. The core–shell particle model considered heat and mass transfer on the shell side and treated the shell as a porous catalytic layer, whereas the core side was a phase change material, in which the latent heat was used to control the reaction temperature.

A full-scale, 2D, packed-bed reactor model was also developed in COMSOL Multiphysics software. The entire reactor behavior was simulated in a transient state, which is based on the core–shell particle properties designed and developed in the particle scale model. The model used a Stanch chemistry model to represent the OCM chemistry, with 10 reaction steps based on the Langmuir–Hinshelwood approach. The chemistry represented nine reaction species including CH_4, CO, CO_2, C_2H_4, C_2H_6, H_2, H_2O, O_2, and N_2. The catalyst chosen for the shell side was a La_2O_3/CaO catalyst, and the core side was decided to be an alloy with cheap metal combinations. The target phase change temperature was 700°C.

Both the reactor and particle models are applicable to any chemical process. The models were not specific to an OCM catalyst's core–shell particles. Particle size and material properties could be changed through a user interface. Moreover, the reactor dimensions could be varied from a lab-scale to prepilot-scale design.

Two possible core–shell catalyst designs were suggested with compositions of Cu(74)–19Zn–7Si and Cu(56)–27Si–17Mg.The shell region was composed of La_2O_3/CaO catalyst. The suggested core–shell particles had a core diameter of 200 μm and a shell diameter of 200 μm. The results indicated that the new catalyst design allowed temperature control at the reactor bed scale as well as the particle scale.

This project leveraged modeling capabilities by introducing new features in packed-bed reactor modeling. Most packed-bed reactor models treat the reactor bed as a porous media, and individual particles are not modeled. The gas phase and solid phase are modeled in continuum such that no heat and mass transfer limitations exist in the particle scale. This assumption limits the applicability of the reactor model to the outside of an isothermal lab-scale design. The reactor model in this project was based on noncontinuum models that considered gas-phase and solid catalyst particles explicitly, which were in turn coupled to model the exchange of interphase transport variables at the fluid–pellet interface. This superior model was capable of incorporating the heat and mass transfer limitations into the particle scale if any existed. Furthermore, coupling a phase change phenomenon is a very challenging problem when combined with

the noncontinuous model. The reactor model developed in this project is certainly the first of its kind and can be applied to different heterogenous catalytic reactions. Furthermore, this method can also be considered for thermal storage media problems. The particle design analogy in this project is an example of how the numerical model can be used for the design of advanced catalyst materials.

MATERIALS SCIENCE AND TECHNOLOGY DIVISION

11007: Novel Environmental Barrier Coatings for Efficient Turbine Engines

D. Sulejmanovic, M. Ridley, H. Wang, A. Rogers

Project Description

This project aimed to develop new environmental barrier coatings (EBCs) through the synthesis of novel doped Y disilicates stabilized in the desired β-phase (β-$Y_2Si_2O_7$) for use in high-temperature turbine engines. Fundamentally, increased fuel burn temperatures (up to 1,700°C) in turbine engines result in greater expansion of the gases and thus increased efficiency or thrust. The SiC-based ceramic matrix composite (CMC) materials are required for achieving higher fuel burn temperatures above the melting points of traditional superalloys, yet CMCs can only be implemented in service with an EBC topcoat to mitigate steam reacting with the SiC. The stability of the β-phase of rare earth element disilicates ($RE_2Si_2O_7$, RE = rare earth element) up to approximately 1,800°C and a low coefficient of thermal expansion (CTE), similar to SiC substrate material, make the β-phase desirable for EBC materials. The β-phase only exists for relatively smaller rare earth element cations (Sc, Lu, Yb, Er, and Y), where Sc, Lu, Yb, and Er are costly and Sc has an undesirable CTE. The compound $Y_2Si_2O_7$ (YDS) is desirable because of its low CTE and higher natural abundance, yet it has multiple undesirable polymorphs between 1,000°C and 1,800°C. A common method for phase stabilization involves introducing multiple rare earth elements in solid solution, yet this method is costly and further increases US reliance on rare earth mineral supplies. This project targeted stabilizing β-$Y_2Si_2O_7$ with two novel cost-effective approaches: (1) cation doping into the Si site [e.g., $Y_2(Si_{2-x}Ge_x)_2O_7$] and (2) aliovalent cation substitutions in the rare earth and Si sites [e.g., $(Y^{3+}_{2-x}Mg^{2+}_x)_2(Si^{4+}_{2-x}P^{5+}_x)_2O_7$]. High-temperature (up to 1,650°C) solid-state synthesis was used to produce polycrystalline solid solution powders. This project introduces a new area of research for low-cost and high-efficiency EBCs, which will encourage faster adoption and implementation of next-generation SiC CMCs in turbines for both aerospace and energy sectors.

Mission Relevance

This project helped to advance the DOE mission by enabling technologies for more efficient turbine engines to reduce fuel consumption in the transportation industry.

Results and Accomplishments

The first approach to synthesis of the stabilized β-$Y_2Si_2O_7$ phase was to dope Ge^{4+} cations in place of Si^{4+}. The initial reactions targeted 10% doping. X-ray diffraction (XRD) of the initial products was performed. This work synthesized the YDS and $Y_2Ge_2O_7$ phases separately and combined them subsequently in a 90-to-10 ratio, respectively. The XRD results showed successful synthesis of YDS and $Y_2Ge_2O_7$ phases. The synthesis of $Y_2Si_{1.8}Ge_{0.2}O_7$ (10% Ge-doped $Y_2Si_2O_7$) resulted in XRD peaks corresponding to phase-pure γ-YDS. The synthesis of phase-pure Ge-doped disilicate is a very encouraging result, but further synthesis parameters need to be tested to see if the Ge-doped β-YDS phase can be synthesized. Based on literature reports on pure rare earth element–doped YDS phases, higher synthesis temperatures (1,800°C) may be needed to obtain the β-YDS phase.

The second part of this project dealt with the synthesis of the β-$Y_2Si_2O_7$ phase by doping into both Y and Si sites by aliovalent doping. The first attempt was the synthesis of $Y_{1.95}Ca_{0.05}Si_{0.05}P_{0.05}O_7$ composition. XRD patterns show the 4 h and 8 h heating attempts. The XRD patterns of the targeted compositions indicate incomplete reactions after 4 and 8 h of heating at 1,500°C. Additionally, a Y_2SiO_5 pattern can be seen after 4 h heating, but the same peaks cannot be detected after 8 h heating, which indicates the consumption of the intermediate phase. However, the formation of the targeted phase, $Y_{1.95}Ca_{0.05}Si_{0.05}P_{0.05}O_7$, cannot be confirmed yet. Longer heating and higher temperatures (1,800°C) may be necessary to improve the purity of these powders.

11252: Steel Compatibility with Beryllides in FLiBe

B. Pint, D. Sulejmanovic

Project Description

This project aimed to evaluate the effect of beryllides and H on the compatibility of reduced-activation ferritic–martensitic (RAFM) steels with molten FLiBe (2LiF–BeF_2) at 550°C–750°C. The use of FLiBe as a liquid tritium breeding concept for fusion energy is of renewed interest to US companies seeking to commercialize fusion as a clean energy source. One series of static capsule experiments was conducted using Mo capsules, and RAFM steel F82H was compared with type 316H stainless steel, which was previously tested in similar experiments. Posttest analysis of the FLiBe salt confirmed that limited Cr and Fe dissolution occurred, and the concentrations did not increase with temperature. Specimens of $Be_{12}Ti$–Be were obtained from Stony Brook University, and commercial Be_2C was obtained for exposure in FLiBe. However, it was not possible to remove residual salt from the specimens after exposure to accurately determine the mass change in the salt at 650°C. Capsule experiments were added for V–4Cr–4Ti and monolithic SiC as a surrogate for SiC/SiC composites. Both showed good compatibility with FLiBe at 650°C.

Mission Relevance

This project helped advance the DOE and ORNL missions by working toward commercializing fusion energy as a clean energy source. Furthermore, FLiBe is a potential liquid breeding blanket concept for fusion reactors, but the understanding of FLiBe compatibility with fusion-relevant materials is still limited. The results from this project demonstrate the dissolution reactions expected as a function of operating temperature.

Results and Accomplishments

Characterization of the salt after exposures of steel specimens showed that relatively low levels of Fe and Cr remained in the salt, and the concentrations did not increase with temperature. A journal manuscript based on these results is in progress.

This project measured specimen mass change after 500 h exposures in commercial FLiBe as a function of temperature and compared Be contents in $Be_{12}Ti$ at 650°C. Coupons (~6 × 12 × 1.5 mm) were welded inside low-carbon arc cast Mo capsules (25 mm outer diameter × 100 mm tall × 1.2 mm wall) with ~28 g of commercially purified FLiBe salt purchased from Kairos Power. Previous fission-focused studies exposed type 316H stainless steel (68 wt % Fe–16.5Cr–10.4Ni–1.9Mo–1.5Mn–0.3Si–0.4Cu–0.034C) in FLiBe, so specimens were included in this study for reference. The mass losses because of dissolution were similar for 316H and RAFM steel F82H (88.9 wt % Fe–8.1Cr–1.8W–0.45Mn–0.20V–0.09Ta–0.08Si–0.06Ni–0.02Al–0.10C–0.01N). At 650°C, three F82H specimens were exposed in three separate Mo capsules to confirm the reproducibility. A potential interaction occurred between Fe and Mo that increased the mass loss, especially at 650°C and 750°C. However, for this initial screening, Mo capsules were a standard choice to contain the salt. Results in the literature suggested that vanadium might react with fluoride salts. However, observed interactions could be due to O_2 or H_2O salt contaminants. The small mass gain for V–4Cr–4Ti (0.03 mg/cm^2) suggested good compatibility. Likewise, high-purity chemical vapor deposition SiC showed a smaller mass loss than that of the steel specimens.

The mass gain for the $Be_{12}Ti$–Be specimens (~12 mm diameter × 3 mm thick) was measured. However, no method was found to remove residual salt from the specimens after exposure, so the mass gains reflect residual salt attached to the coupons and did not determine if any reaction occurred with the salt. The next experiment was to expose the $Be_{12}Ti$–Be specimens with F82H specimens to determine whether any interaction occurred. These capsules were prepared but could not be completed during the performance period because of the late arrival of the salt. Likewise, the second phase of testing was to include F82H specimens loaded with H_2, but these tests also were not completed.

After the exposure, the specimens were characterized to determine the extent of the attack. Specimens were imaged using scanning electron microscopy, and chemical changes were studied using energy-dispersive x-ray spectroscopy maps and line profiles. The data were collected using a ZEISS model GeminiSEM 460. Typical of most observations in halide salts, selective attack of Cr in the salt occurred, and a rough surface suggested that Fe dissolution also occurred. To quantify the dissolution, Fe and Cr concentrations were measured in commercial FLiBe salt as a function of exposure temperature after 500 h exposures with F82H and 316H specimens. Based on the mass losses, one would expect more Fe and Cr in the salt with increasing temperature. However, the Fe–Mo interaction resulted in less Fe in the salt and low levels of Cr in each of the tests. This finding agrees with prior experiments. The measured Cr and Fe data for the three F82H capsules run at 650°C showed remarkable reproducibility.

11271: Chiral Materials for Next Generation Quantum Transduction

M. Brahlek, P. Ganesh, D. Lingerfelt, J. Serafini

Project Description

This project sought to synthesize and probe new materials that would facilitate a long-distance quantum network. To realize a so-called *quantum internet*, optoelectronic-based networks were needed. These networks distributed quantum information to classical and quantum computing systems, which offered capabilities to transmit data that were encoded by the fundamental rules of quantum mechanics. This challenge was directly addressed with a materials redesign. To realize this goal required overcoming key materials challenges, including predicting new chiral materials and synthesizing candidates while minimizing deleterious defects that would ultimately cause signal dissipation during transmission, endangering the fidelity in quantum measurements and manipulation steps.

Mission Relevance

This project delivered preliminary data on new materials that are necessary to realize new applications in microelectronics and quantum science.

Results and Accomplishments

The key results of the project were making headway in the growth of several materials that exhibit chiral properties as well as work on the theorical side to better find and predict chiral materials. The key findings of this work were the ability to synthesis chiral material such as Te and materials that exhibit chiral spin–momentum textures such as MnTe on several quantum platforms. This project synthesized chiral Te on graphene, as well as the novel magnetic platform MnTe. In this work, the data showed high-quality films in which the chiral properties are clearly visible in the transmission electron microscopy, addressing a key project objective, which was to understand how to synthesize chiral materials and integrate them with other materials at atomically precise interfaces. The MnTe has novel chiral properties in the spin-channels. This project discovered that for MnTe on InP, rich magnetoresistance effects occur from this system, which show multiple atomic force microscopy–derived spin-flop transitions as well as emergent ferromagnetism. As such, this work demonstrates the potential for a tunable system with rich magnetic structure that derives from the spin–momentum locking. This project integrated structurally chiral Te with MnTe, finding that both MnTe and Te can be integrated with the unconventional superconducting system FeTe. This outcome is relevant to a number of questions regarding how superconductivity is affected by structural chirality through the proximity effect. Mixing the unusual chiral spin–moment textures in MnTe with superconductivity at these interfaces may give rise to unconventional effects. For example, electronically, topological superconductors are proposed to exhibit novel chiral spin–triplet pairing, and structurally chiral superconductors give rise to exotic properties such as strong magnetoelectric coupling with strong spin-polarized carriers and extremely large upper critical fields.

11286: First-Principles Spin-Phonon Coupling: Spin and Lattice Transport

L. Lindsay, R. Fishman, X. Li, R. Juneja

Project Description

Theoretical and numerical tools for quantitative prediction of spin–phonon couplings and subsequent material properties are not fully developed. This challenge limits the ability to understand lattice and spin behaviors, predict transport properties, and tune material behaviors in novel quantum magnets and other spin-based materials for thermal management, spintronics, and quantum information applications. In this project, the team aimed to develop first principles–based numerical tools for modeling spin–phonon interactions from density functional theory (DFT) methods for a wide variety of magnetic systems of interest to DOE. With accurate descriptions of spin–phonon couplings, algorithms can be developed to quantify material behaviors of quantum magnets, including spin and lattice thermal transport. The team leveraged its expertise in first principles–based calculations of phonons and magnons to build transport descriptors based on quantum perturbation theory. Ground-state energies and forces of magnetic systems were calculated via standard DFT packages (i.e., Vienna Ab initio Simulation Package and Quantum Espresso). Spin–phonon coupling matrix elements were built from large numbers of perturbed supercells and served as inputs to quantum perturbation theory to describe spin–phonon interactions. Transport phenomena were then calculated from Boltzmann transport theory with these interactions as resistance.

Mission Relevance

This work addresses a variety of scientific and applications needs defined in DOE BRN reports in Microelectronics (BRN-Micro), Opportunities for basic research for Next-Generation Quantum Systems (BRN-NextGenQS), and Quantum Materials for Energy Relevant Technology (BRN-QMEnergy). More specifically, the work here highlights pathways for manipulating spin–phonon interactions for enhanced design of coherent quantum spin systems. Thermal management is a high priority in the design of novel microelectronics architectures, and understanding spin–phonon coupling thermal resistance will guide the design of novel magnetic memory systems. The team's numerical methods will affect multiple BES MSE efforts and will support programs in the Quantum Science Center for which spin coherence and manipulation are critical, particularly in reducing thermal vibrational interference. Additionally, this work closely couples with research programs using neutron scattering experiments to probe phonon and magnon excitations.

Results and Accomplishments

The team developed numerical algorithms to describe collinear magnetic excitations, magnon Berry curvature, magnon orbital angular momentum, and magnon surface conducting states. The team explored possibilities of employing DFT and Wannier mapping methods for deriving spin–phonon coupling terms. The team developed quantum perturbative methods to describe scattering and lifetimes for phonon and magnon transport predictions.

NUCLEAR ENERGY AND FUEL CYCLE DIVISION

10991: Bayesian Neural Networks for Scientific Discovery in Nuclear Data

G. Arbanas, V. Reshniak, M. Pigni, J. Brown, D. Wiarda, J. McDonnell, K. Roche, H. Abdel-Khalik, V. Sobes

Project Description

This project was a first-of-its-kind application of machine learning (ML) optimization techniques for the discovery of scientific insights from extant databases of measured nucleon–nucleus cross sections, which may yield improved evaluated nuclear data for accurate neutron transport simulations of nuclear engineering applications. This project facilitated solutions to nuclear energy challenges by improving nuclear data evaluations to improve the efficiency and accuracy of the nuclear reactor design process while minimizing design uncertainties.

Mission Relevance

This project is the first to apply an ML method to research nuclear reactions required for nuclear data evaluations used for neutron transport simulations of nuclear energy reactors, which are inherently free of greenhouse gas emissions.

Results and Accomplishments

This project delivered a prototype ML tool[1] for investigating novel parameterizations of a nucleon–nucleus optical potential model (OMP) that is expected to improve the conventional Woods–Saxon parameterization (historically used in the 1 keV to 200 MeV nucleon–nucleus energy region based on intuitive parameterizations of phenomenological OMPs), thereby effectively capturing the observed experimental cross-section data for stable nuclei. However, when extrapolating conventional models for nuclides with limited or unavailable data, global models that parameterize the OMP across the entire chart of nuclides must be estimated. Unfortunately, this approach results in a noticeable reduction in predictive power because of the limited expressive power of the Woods–Saxon parameterization. To address this limitation, this project developed a deep learning approach for parameterizing OMPs and developed a training algorithm that combines PyTorch models for the potentials and the black box ECIS nucleon–nucleus scattering solution code used to calculate OMP cross sections. The team trained its ML OMP models by using data from evaluated and experimental nuclear data libraries such as the TALYS Evaluated Nuclear Data Library (TENDL) and EXFOR. The preliminary results demonstrated an improved agreement with ^{93}Nb total cross section data relative to conventional OMP parameterizations.

11189: Integration of a Direct Air Capture System in the Thermal Balance of An Existing Nuclear Power Station

E. Popov, V. Kumar, A. Sircar

Project Description

The purpose of this project was to conduct a front-end engineering design study of an advanced direct air capture (DAC) system that is powered by nuclear energy and capable of the removal from the air and long-term storage of a minimum of 5,000 t/year net CO_2. A low-temperature DAC system powered by an operating nuclear power plant (the Watts Bar Nuclear Plant, TVA's Westinghouse-based four-loop power plant located near Spring City, Tennessee) was assessed. A techno-economic analysis, which was performed to establish a common basis for evaluating carbon capture technology, was useful in identifying possible areas of technological improvement that can help bring down the cost. A balance of

[1] ORNL. "Autotalys." https://code.ornl.gov/vre/autotalys/.

plant was developed in Modelica[2] for Watts Bar and was coupled to a black-box model of the Global Thermostat low-temperature DAC system.[3]

The costing of the system used the life cycle analysis concepts developed by Terlouw et al.,[4] in which the "process unit" of a DAC system comprises an absorber (which also acts as the regenerator for CO_2 release) and fans to force air into the absorber. In addition to the process unit, an entire DAC system constitutes the hall that houses all control and monitoring systems and storage boxes that store the compressed CO_2 until it is transported to an off-site facility. The method used for costing of the DAC system followed a procedure used to cost postcombustion capture that was scaled down for 5,000 t/year of CO_2 removal.

The DAC costing follows the same principles as the capital cost analysis. DAC has very low output, which causes the unit cost to go up. Building such low-capacity plants is not cost-efficient and should be avoided. Powering the DAC fully from the nuclear plant does not affect plant efficiency because the DAC has low capacity. Significant expense would be saved, but the relative labor and administration costs would increase. Remaining factors (e.g., taxation, plant life) are standard for a chemical plant.

Mission Relevance

This project evaluated (1) the feasibility and energy efficiency of DAC powered by nuclear energy and (2) the environmental, economic, and social aspects within an integral front-end engineering design study. The DAC system design is a mature technology. Carbon capture technologies are being actively researched around the world to meet net-zero goals by 2050. Economically feasible deployment of these technologies at the industrial scale is important for early adoption. Using a zero-carbon energy source such as nuclear power to remove CO_2 from the atmosphere by DAC will help meet the world's carbon-neutral energy goal. Using nuclear power is expected to reduce the resultant gross CO_2 yield by 33%.

Results and Accomplishments

The study demonstrated that a system in which DAC is coupled with a nuclear power plant can be leveraged to remove up to 100,000 t of CO_2 annually without significantly affecting the balance of plant. A life cycle analysis was carried out for the coupled system. Using energy from a nuclear power plant with a projected 20-year plant lifetime to meet the thermal and electricity needs of the DAC system results in a relatively small climate change impact: 69.9 kg of CO_2 would be released to remove 1 t of CO_2 from the atmosphere. A nuclear-powered DAC system would have a 93.0% carbon removal efficiency (i.e., the net amount of CO_2 removed over the total amount of CO_2 captured[5]). This finding is comparable with the carbon efficiencies obtained from renewable sources to meet the energy demands of the low-temperature DAC system. The contributions to global warming of the energy demands for operation and storage combined are comparable with the contributions by the DAC infrastructure requirements and adsorbent consumption (approximately 38%). This result indicates that the total global warming potential could be brought down considerably with further improvements in the DAC design and adsorbent performance.

[2] "Modelica." Modelica Association. Accessed December 14, 2023. https://modelica.org/.
[3] "The Air Carbon Solution." Global Thermostat. Accessed December 14, 2023. https://www.globalthermostat.com/.
[4] T. Terlouw et. al. "Life Cycle Assessment of Direct Air Carbon Capture and Storage with Low-Carbon Energy Sources." *Environmental Science and Technology* 55, 16, 2021, 11397–11411. DOI: 10.1021/acs.est.1c03263.
[5] S. Deutz and A. Bardow. "Life-Cycle Assessment of an Industrial Direct Air Capture Process Based on Temperature Vacuum Swing Adsorption." *Nature Energy* 6, 2, 2021, 203–213. DOI: 10.1038/s41560-020-00771-9

11223: Neural Network Surrogate Model for Radiation Damage in Concrete

A. Cheniour, M. Lupo Pasini, I. Greenquist, Y. Le Pape

Project Description

The concrete biological shield (CBS) in light-water reactors is subjected over time to high levels of neutron radiation, which causes significant damage to its inner portion, resulting in the degradation of the concrete's shielding properties. Ongoing efforts to predict the extent of damage are based on separate mesoscale (a few centimeters) and CBS structural (a few meters) models. Ideally, the two models would inform each other by providing information on the local damage and expansion under irradiation at the mesoscale and on the neutron fluence, temperature, and mechanical load at the structural scale at each location and time considered. However, the coupling is computationally intensive and impractical because of the number of mesoscale simulations needed to inform the entire CBS domain. The recent development in neural network–based surrogate models for computational mechanics applications enables a significant increase in computational speed of multiscale models with high accuracy. This project developed an inexpensive surrogate model by training a graph convolutional neural network (GCNN) on the finite element mesoscale model subjected to neutron irradiation and mechanical load.

Mission Relevance

This work employed advanced DOE simulation tools and high-performance computing resources and established a methodology to significantly reduce the computational cost of fully coupled multiscale models while maintaining the desired accuracy. This approach provides an important advancement in the application of GCNN models in computational mechanics as well as in the modeling of irradiated concrete. The broader outcome of this work is its generalizability to other problems relevant to DOE's mission. For instance, once such problem is linking mesoscale fuel simulations to engineering-scale fuel performance predictions to account for local effects such as radiation damage and fission gas behavior, which are highly dependent on time and the radial position in the fuel pellet.

Results and Accomplishments

The specific goal of this project was to provide a computationally efficient and high-accuracy GCNN surrogate model that emulates the mechanical behavior of irradiated concrete at the mesoscale to inform a structural model of the CBS on local and time-dependent expansion and damage.

This project successfully built a graph neural network surrogate model that predicted the average expansion and damage of concrete as a function of temperature, exposure to radiation, intensity of radiation, and time. Because of the large volume of finite element data used for the training, the training of the surrogate model has been scaled on Summit using up to 40 graphics processing units. Moreover, a binder has been developed to integrate the trained HydraGNN surrogate model within the C++ finite-element code MOOSE (i.e., Multiphysics Object-Oriented Simulation Environment) for accelerated prediction of concrete damage in nuclear reactor shielding applications. The Python binder has been released as open-source with technology transfer.[6]

11242: Optimal Chloride Salt Mixture for a Fusion Blanket

N. Goth, L. Scott, T. Ghaddar, J. Brown, P. Britt

Project Description

The purpose of this work was to investigate novel liquid blanket materials that could provide self-sustaining operation, drawing on experience from research on molten salts used for advanced fission

[6] A. Cheniour, I. Greenquist, and M. Lupo Pasini. *MOOSE-Python Binder*. Oak Ridge, Tennessee: Oak Ridge National Laboratory, 2023. https://www.osti.gov/biblio/2006611

reactors, concentrated solar, and thermal energy storage. The hypothesis when proposing this research was that there could be chloride-based blanket designs that can exceed the tritium breeding ratios of (FLiBe) molten salt blankets while reducing the use of Be (FLiBe), avoiding the generation of HF (FLiBe), and minimizing magnetohydrodynamic (MHD)-perturbed flow fields (PbLi). The fastest and most cost-effective path to deploying fusion breeder blankets could be from maximizing the synergistic technological overlap between fusion, fission, concentrated solar, and thermal energy storage industries.

The goal of this research was to systematically identify chloride-based salts with desirable economic, neutronic, thermophysical, and thermochemical properties for use as a liquid fusion breeder blanket and then compare such plausible salts with state-of-the-art FLiBe molten salt. If proven to yield sufficient tritium production, a chloride salt blanket has the potential to avoid the primary disadvantages of FLiBe (presence of Be toxicity and generation of HF) and PbLi (MHD affecting fluid flow and heat removal).

This work generated a near-complete list of salts by searching vendor inventories. Plausible combinations were generated by iterating through permutations of unary salts to yield 3,081 binary, 79,079 ternary, and 1,502,501 quaternary salts. The lists were expanded to include weight fractions with a 10% step size. The inclusion of this dimension generated 9 compositions for each binary salt and 36 for each ternary salt.

A multilevel serial filtering process was applied. This set of filters was loosely designed to minimize computational cost. This systematic downselection of the most promising chloride-based salts focuses on economic, neutronic, thermophysical, and thermochemical parameters. After filtering, the most promising chloride salts were investigated using transport and depletion sequences within the SCALE suite of software products. Finally, from this dataset, the best performing chloride salts were compared with FLiBe.

Mission Relevance

Deuterium-tritium fusion reactors cannot operate for a significant period without a closed tritium fuel cycle according to a recent NAS report[7] on bringing fusion reactors to the United States' electrical grid. This fact places breeder blankets as one of the foundational systems for self-sustained fusion reactor operation. This research aligns with the DOE's bold decadal vision for commercial fusion deployment by seeking to explore blanket configurations that have significant overlap with existing research and industries to accelerate the technology readiness level of a liquid breeder blanket.

Results and Accomplishments

After using the previously mentioned properties as filters, low-, medium-, and high-cost neutronic simulations were performed to quantify tritium production on reactor geometries.

The top-performing chloride salts were compared with FLiBe as the benchmark. Over 30 binary and 20 ternary compositions were identified with superior tritium production. Although tritium production is the highest-priority metric, a balance must also be achieved with other economic, thermophysical, and thermochemical parameters. The most promising binary salts were scored and ranked using tritium production, liquidus temperature, price, toxicity, ionic states, redox potential, activation potential, fluid flow, and heat transfer metrics.

Several aspects of chloride salt performance relative to FLiBe are highlighted. Several compositions were found with higher-than-FLiBe tritium production potential. Chloride salts have been identified with higher-than-FLiBe tritium production without Be or Pb. Some chloride salts can exist in multiple ionization states, which complicates blanket chemistry. Activation results suggest that chloride salts have larger decay heat values than FLiBe. Figures of merit for fluid flow and heat transfer define chloride salts as less desirable than FLiBe primarily because of lower specific heat capacities. The $MgCl_2$ and $CaCl_2$ salts are already used in the halide slagging step of the pyrometallurgical process. Also, LiCl–KCl is used

[7] R. J. Hawryluk et al. *Bringing Fusion to the US Grid*. The National Academies of Science Engineering and Medicine Public Briefing, February, 17, 2021.

in the electrorefining step of the pyrometallurgical process, so significant operational experience and thermophysical and thermochemical properties exist.

A technical report[8] has been published capturing the approach and results of this research.

[8] N. Goth, J. Brown, T. Ghaddar, and L. Scott. *Optimal Chloride Salt Mixture for a Fusion Blanket*. ORNL/TM-2023/3052. Oak Ridge, Tennessee: Oak Ridge National Laboratory, 2024. DOI: 10.2172/2282960

PHYSICS DIVISION

11258: Ring Resonators for Imaging of Gravitational Fields

M. Willis, J. Serafini, M. Febbraro

Project Description

This project evaluated the feasibility and sensitivity of microelectromechanical (MEMS) ring-resonator arrays for imaging of gravitational fields within nuclear nonproliferation and high-energy physics mission spaces. Gravimetry is used in a wide range of applications, including petroleum exploration, geophysical mapping, archaeology, mineral exploration, basic science, nuclear security, military applications, and planetary exploration. These applications are possible, in part, because of the ability of gravimeters to measure mass distributions via local gravitational fields. Existing gravimeters suffer from a few challenges, including sensitivity, background noise, and cost. These challenges make imaging of gravitational fields economically unfeasible. Recent advances in MEMS technology have produced accelerometers that can be used to measure relative gravitational field changes (i.e., mass distributions) with sensitivities comparable with commercial gravimeters at a fraction of the mass and cost. Additionally, the recent achievements in manufacturing high-Q ring resonators serves as a scalable readout scheme for arrays of MEMS accelerometers. By combining low-cost MEMS accelerometer arrays with multiplex ring resonator–based readouts, this project can improve detection sensitivity by canceling out common mode noise (i.e., an array of coupled sensors) needed for gravitational field imaging applications.

Mission Relevance

Gravimetry offers an advantageous approach to passive measurement techniques because it can measure relative mass distributions at a standoff distance. As a result, gravimetry has the potential to perform nondestructive assessment of the mass distribution of nuclear or accompanying materials in containers without opening them to adjudicate incomplete records, assess safety, perform nuclear material accounting, or provide quality assurance. Within DOE HEP, this technology offers a detection modality for future dark matter searches. Gravimetry enables measurements of the one fundamental force; it is known that dark matter interacts with gravity. For future dark matter searches to be viable, large arrays of accelerometers (relative gravitometers) will be required, for which a viable technology has yet to be found. This work also shares synergy with the National Microelectronics Initiative through advances in photonic integrated circuits.

Results and Accomplishments

This project aimed to design, build, and test ring resonators and interferometer devices by removing SiO_2 above, around, and underneath Si waveguides. The etch regions (windows) were spaced out so that portions of the waveguide were not anchored to the substrate, allowing the waveguide to interact with neighboring material and have strain caused by external stimuli subsequently change the optical mode's effective index. Under acceleration, the ring can become deformed and change the index of refraction, causing the resonance wavelength to shift (and allow for gravity to be measured).

To compare chip performance, the team planned measurements with a commercial gravity meter. Laboratory measurements showed that commercial gravimeters can measure relative gravity owing to distance and mass changes as small as 5 kg. Field measurements were also performed for multiple line scans at a pillar-and-room mine type that allowed the team to estimate gravity anomalies as a function of depth. Laboratory and field gravimetry measurements proved to be a useful tool for measuring relative gravity with a commercial unit. This unit, however, does not scale well, and a solution using the resonators will hopefully allow for larger areas to be covered and increased sensitivities to be achieved. Additional comparative measurements will need to be performed between the commercial unit and the custom SiO_2 etched resonators to determine sensitivities and capabilities of the new devices.

RESEARCH ACCELERATOR DIVISION

11008: Condition Monitoring and Advanced Packaging for Accelerator Modulator and Electric Vehicle Motor Drive Insulated Gate Bipolar Transistor Devices

C. Pappas, D. Anderson, A. Bullman, S. Harave, O. Onar, J. Walden, J. Wilkins, S. Chowdhury

Project Description

Insulated gate bipolar transistors (IGBTs) are widely used in power electronics applications, including in motor drives and battery charging for electric vehicles and in power supplies and pulsed power for particle accelerators. Although generally very reliable, they are susceptible to degradation caused by wire bond and solder fatigue and catastrophic failure in certain fault conditions. The purpose of this proposal was to investigate (1) using condition monitoring techniques to warn of the degradation of IGBT modules and schedule preventative maintenance of the high voltage converter modulators (HVCMs) at the Spallation Neutron Source (SNS) and (2) the internal structure of the IGBT connections to improve IGBT reliability during extreme fault conditions.

Mission Relevance

This work is relevant to future modulators and inverters as higher switching frequencies, larger di/dt IGBT operation, and increased system reliability become major design objectives in the systems where these devices are deployed. Because the emphasis in the accelerator and electric vehicle power electronics is shifting to improved reliability, this work has relevance to retrofitting into existing systems and to consideration for new designs.

Results and Accomplishments

For the condition monitor project scope, 36 articles were reviewed to identify the condition monitor parameters most likely to lead to improved reliability at SNS. The team decided to investigate 5 out of the 20 that were identified in the literature review: junction temperature, collector-to-emitter voltage, collector current, gate-to-emitter voltage, and gate current. A test circuit was designed and fabricated to allow for high-power measurements of the parameters, and an environmental chamber, thermal imaging camera, and suitable voltage and current probes were used. Baseline measurements were made on a new device over a range of temperatures.

For the device packaging part of the project, a device that was damaged in service at SNS had the case and silicone potting compound removed the expose the silicone dies. A 3D mechanical model of the package was then created and imported into Ansys Maxwell for electromagnetic field calculations. Both the overall device inductance and sharing of current between dies and bond wires could be improved by adding a power collector connection and the removal of a cut in the power emitter connections. A circuit model of a raft was also developed; however, it was found that Ansys Maxwell was unable to accurately calculate the inductance matrices.

SUMMARIES OF PROJECTS SUPPORTED BY THE STRATEGIC HIRE PROGRAM

BIOSCIENCES DIVISION

10493: Quantitative Systems Modeling for Physiology-Informed Molecular Design

B. Akpa, N. English

Project Description

The purpose of this project was to develop mechanistic models to support molecular design in the specific use case of drug discovery. In drug discovery, molecular interactions are intended to modulate the biological pathways underlying human physiology in well-defined ways, but the availability of physiological data is limited by multiple factors, including ethical factors. This project developed a mathematical model that, given physicochemical properties of a virtual chemical agent, predicted the chemical agent's time-dependent distribution within the human body (i.e., pharmacokinetics) and its effect on physiological function (i.e., pharmacodynamics) with associated quantification of uncertainty. Outputs from the model served as biologically informed design criteria for an artificial intelligence (AI) platform that performs generative molecular design, which is currently in development by members of the Accelerating Therapeutics for Opportunities in Medicine Consortium. By iteratively integrating these organism-level models with the pipelines for molecular property prediction and generative molecular design developed by collaborators, this project enabled molecular design for the correction of emergent physiological dysfunction.

Mission Relevance

The DOE BER mission aims to achieve predictive understanding of biological systems from genes to ecosystems. Integral to this mission is a desire to render this knowledge actionable so that biosystems outcomes can be engineered in support of energy and security needs. Integrating predictive mechanistic models with generative design approaches can deliver the capacity to engineer biosystems-level outcomes via molecular design. In this context, mechanistic models provide surrogate data to reflect causal links between molecular events and systems-level phenomena, which is important when data at the systems level are difficult or impossible to obtain at a sufficient scale to enable data-driven models.

This project demonstrated how mechanistic systems models can serve as the link between molecular properties and biological function. Success was measured by the project demonstrating a capacity to perform a presynthesis, multifactorial optimization of drug therapeutic value that explicitly accounts for human outcomes.

Results and Accomplishments

The overall goal of this project was to enable presynthesis discovery of high–therapeutic value molecular candidates by integrating human systems models into generative design workflows. To achieve this goal, the following challenges had to be overcome: challenges associated with (1) identifying suitable models for predicting drug efficacy, (2) evaluating model validity, (3) quantifying uncertainty, and (4) summarizing model output to allow interfacing with a generative AI model via an optimization algorithm.

This project successfully integrated generative AI, machine learning, and dynamic systems models in a future-proof, scalable computing workflow to enable inverse design of molecules. Incorporating systems models that projected emergent biology from molecular perturbations allowed these workflows to select molecules expected to drive defined, emergent outcomes. Accordingly, this work demonstrated the distinct molecule set identified by invoking dynamic systems outcomes as molecular design criteria. The results indicated the resources that might otherwise be wasted on downstream development of molecules initially optimized for molecular properties alone.

In the process of meeting this goal, this project demonstrated (1) the value of adapting the assumptions within mathematical models to minimize sensitivity to poorly defined input parameters across broad chemical spaces and (2) the role dynamic systems outcomes can play in facilitating robust molecule rankings in the face of parameter uncertainties. In both cases, this project designed the systems model to be tolerant of anticipated limitations of the machine learning models that provide input parameters.

In short, the project has yielded a new capability: a scalable, inverse design workflow as an exemplar for generative molecular design that can drive desired emergent properties of biological systems (or, indeed, other complex systems).

10734: Relationships Between Plant Structure and Dielectric Permittivity for Soft Actuators, Energy Storage, Remote Sensing, and Non-Destructive Characterization

S. Bhagia

Project Description

Research to determine the dielectric properties of plant cell wall components is a burgeoning field of study that can have profound benefits for energy storage, plant diagnostics, and remote sensing. Complex permittivity measurements of plant biomass over a broad frequency range will yield valuable information about biomass structure at multiple length scales. It is anticipated that data from such studies will be relevant to the understanding of dielectric permittivity behavior in plant systems and will facilitate the development of novel material applications. In past studies, permittivity measurements were carried out on isolated celluloses and involved a comparison of a few plant species. No studies have clearly established relationships between permittivity and biomass parameters such as cellulose crystallinity; acidic groups of hemicellulose; hydroxyl, methoxy, and carboxylic groups of lignin; or ratios of syringyl to guaiacyl lignin.

The purpose of this project was to utilize ORNL's genome-wide association studies (GWAS) of a *Populus trichocarpa* population in the first large-scale broadband dielectric spectroscopy (BDS) study to find out how the structure and composition of biomass polymers (cellulose, hemicellulose, and lignin) affect permittivity over a wide frequency range (10^{-2} to 10^{10} Hz). It was accomplished by isolating 15 to 20 cellulose, hemicellulose, and lignin GWAS samples and thoroughly characterizing them by nuclear magnetic resonance (NMR) and infrared (IR) spectroscopy. BDS was then carried out over a range of temperatures (–60°C to 120°C) on bone-dry vs. equilibrium moisture–containing samples to find correlations and assign specific dielectric relaxation frequencies to permanent dipoles that occurred in individual biomass components. BDS measurements were made at room temperature on bone-dry and equilibrium moisture–containing samples from the ORNL GWAS poplars. Statistical and chemometric techniques were applied to lignin data obtained from spectroscopy measurements (NMR, Fourier transform, IR, and near-IR). The results established a solid foundation for determining the relationship between biomass structure and permittivity over a broad frequency range.

Mission Relevance

The project established new and unique expertise in the dielectric properties of biomass, which could ultimately be used to address DOE's BETO and EERE AMMTO goals in decarbonization, advanced manufacturing, and plastic waste mitigation.

Results and Accomplishments

Lignin was artificially synthesized on the surfaces of fibrillated bleached softwood pulp. Pulp containing 85% cellulose and 15% xylan was treated with xylanase to remove xylan from accessible cellulose surfaces. The xylanase-treated pulp retained 10% of the xylan, which was inaccessible by the xylanase enzymes. Lignin was then grown on surfaces of cellulose–xylan and low-xylan cellulose fibers by

reacting with coniferyl alcohol using laccase-based free-radical polymerization. This step was done to (1) determine the lignocellulosic structure in plants by dielectric spectroscopy and related techniques based on atomic force microscopy (AFM) and (2) enable the development of new renewable materials. This research discovered that the lignin content was 8.66% in the cellulose–xylan solids and that the lignin content was 16.2% in low-xylan cellulosic solids. The 46.2% higher lignin content on the removal of accessible xylan shows that hemicellulose is a governing factor for lignin formation in softwood lignocellulose. Therefore, cellulose–xylan, cellulose-inaccessible xylan, cellulose–xylan–lignin, and cellulose-inaccessible xylan–lignin fibers were characterized by several NMR techniques, and AFM-based nanoscale spectroscopy modalities (e.g., IR, Raman, and impedance spectroscopy) were applied to get insights into the structure of model softwood lignocellulose fibers. Additionally, special gel-state 2D NMR techniques were applied to determine the native structure of the fibers.

The new AFM-based, scattering-type, near-field optical microscope with broadband IR spectroscopy functionality was used to study a wild-type poplar and a reduced-pectin mutant of transgenic poplar. The results showed that there were more compositional differences between the wild-type and the mutant in secondary cell wall regions than in the compound middle lamella regions. The ratio of hemicellulose or lignin to cellulose increased in the mutant compared with those in the wild type for the secondary cell walls and the compound middle lamellae. Distinct regional sectors existed within secondary cell walls and the compound middle lamellae with respect to cell composition that appear to be controlled by a periodic change in growth. This kind of nanoscale imaging addresses the need for viewing the distribution of components in biological tissues at the nanoscale level, which was not possible until the development of new quantum cascade lasers and precision in optics.

11185: Towards Mapping Sequence–Activity Relationships of an Entire Microbial Proteome

C. Eckert, W. Alexander, E. T. Prates

Project Description

A fundamental goal of synthetic biology is the development of new paradigms to enable the accelerated engineering of biological systems. This Design, Build, Test, Learn cycle recursively uncovers design rules behind phenotypes of interest and will eventually enable advanced computational approaches for design prediction. Key information for the Design, Build, Test, Learn cycle is sequence-to-activity relationship (SAR) mapping, but historically, elucidation of these relationships has been limited owing to a lack of high-throughput genetic tools with which to efficiently explore the design space for a given component of interest. Advances in DNA synthesis, sequencing technologies, and high-throughput genetic toolkits for a growing variety of microbes have paved the way for massively parallel, genome-scale SAR approaches. Previous work related to this project developed and implemented CRISPR-Cas–based technologies that this project intended to demonstrate as a framework for massively parallel, genome-scale SAR mapping at the amino acid level. This project built on recent work to map essentiality at the amino acid scale in *E. coli*. Because of its developed high-throughput genome editing and transformation tools, *E. coli* is an ideal model system for demonstrating massively parallel, genome-scale libraries for saturation mutagenesis. Implementing these tools in *E. coli* will allow researchers to map these amino acid changes to the protein structure and represent the first steps to mapping the SAR of the entire proteome of a microbe. This framework will enable new engineering strategies with broad implications across synthetic biology and general biotechnology.

Mission Relevance

Mapping the SAR for the entire proteome of a microbe will enable a more in-depth understanding of the underlying mechanisms controlling cellular processes. Additionally, it will enable new engineering strategies with broad implications across microbial species of interest for biotechnological applications.

Results and Accomplishments

Although CRISPR-enabled trackable genome engineering (CREATE) is a technique that this team has used in the past for generating multiplexed, edited libraries under various selective conditions, issues still exist with cells that escape editing. Wild-type cells left in populations can negatively affect the ability to select out causative mutants. The team found that including spike-in plasmid controls solved the problems that the team faced with interpreting selection data and normalizing background. The team also sought to develop a protocol to arrest cells that escape editing to decrease the amount of unedited cells in the population. The team found orders of magnitude of improvement in recovered, edited cells with a standard control by growing transformants on minimal defined media.

To improve the design of guide RNAs (gRNAs) and editing cassettes to avoid unedited cells in the studied populations for deep mutational scanning and site saturation mutagenesis, this work sought to develop bespoke code to produce gRNA libraries to test all possible gRNAs in *E. coli*. Code for producing CREATE cassettes was adapted and updated to produce gRNA-only libraries. Included in this work were modules that were able to find potential off-target sites by excluding spacers that had strong matches at noncanonical protospacer associated motif sites (for *S. pyogenes* Cas9, NAG, and NGA protospacer associated motifs).

To understand design rules for gRNAs and their ability to target the genome, this study generated a library pool containing all possible gRNAs in *E. coli* in a cloning strain. Preliminary analysis was conducted on the 0.5% of gRNAs that strongly enriched in vivo (which indicates a total lack of function) with an eye toward their predicted secondary structure. Many of these gRNAs had highly stable secondary structures, which has been shown to disrupt Cas9 activity in vivo. These data are serving as a training set for an explainable artificial intelligence model to help predict bad gRNA sequences and exclude them from use in genome editing libraries. Using what the team learned from the gRNA targeting experiment, researchers sought to develop bespoke code to produce the deep mutational scanning and site saturation mutagenesis libraries. A library design script was written to produce these libraries. The script was also written to address potential design needs, such as targeting individual mutations in a gene rather than mutating the entire gene start to end.

The team developed a semiautomated workflow to collect protein structural and functional data. The initial design has been completed for the pipeline and simulation with a protein from the *E. coli* central carbon metabolism. The team also designed a comprehensive library of curated and predicted structural and functional data for *E. coli* central carbon metabolism (CCM) proteins to guide these initial models. The team has assembled a comprehensive library of 61 distinct features for each amino acid of *E. coli* CCM proteins. The primary objective is to leverage this rich feature library as a training and test set for a gradient boosting machine, aiming to effectively classify amino acids as either essential or neutral. Notably, this approach has demonstrated success in the past with plants,[1] albeit using a smaller feature library.

[1] M. Gao, D. Nakajima An, J. M. Parks, and J. Skolnick. "AF2Complex Predicts Direct Physical Interactions in Multimeric Proteins with Deep Learning." *Nat. Commun*. 13, 2022, 1–13. DOI: 10.1038/s41467-022-29394-2

CENTER FOR NANOPHASE MATERIALS SCIENCES

10710: Electron, Ion, and Photon Mediated Modification of Two-Dimensional and Quantum Emitter Materials for Precision Manufacturing

S. Randolph

Project Description

Low-dimensional and quantum emitter materials promise notable functional capacity and flexibility, exhibiting unique properties that emerge when they are combined and integrated in unique ways. These materials will likely play a significant role in the development of next-generation sensing and computational platforms. However, the unique morphologies and properties that make 2D and quantum emitter materials so attractive are offset by environmental susceptibilities and sensitivities to degradation and damage. These sensitivities present formidable challenges to integrating these materials into functional devices as prototypes and even more so when attempting to manufacture at scale. If the potential of these promising materials is to be fully realized, reliable fabrication methods that do not compromise valuable material properties must be developed. Within the electronics industry, the role of electron, ion, and photon direct-write nanomanufacturing techniques has traditionally been confined to prototyping, lithography, and failure analysis. However, the accelerated development of these materials is converging with new focused electron and ion beam technologies. This convergence presents opportunities for direct-write processing to play an expanded role in tuning material properties and integrating them into functional architectures in a scalable manner.

The purpose of this project was to cultivate a deeper understanding of the underlying damage mechanisms that can occur with direct-write processing of these materials and to create a suite of strategies for mitigating or exploiting these beam–material interactions in the context of synthesis, device integration, and scaled precision manufacturing. Specifically, the project investigated the fundamental mechanisms by which ion beams introduce defects and dopants to 2D and quantum emitter materials. Controlling the density and spatial distribution of the defects and dopants can shape the functional properties of the materials. The effect of electron beam–induced etch (EBIE) chemistries on selectivity during the removal of target 2D materials was examined, ultimately enabling spatially defined atomic layer etching of heterogenous 2D material stacks. The role of underlying surface chemistry and substrate morphology was investigated to exploit local surface termination and strain to alter susceptibility to doping and defect creation.

Mission Relevance

The work in this project was designed to expand expertise in ion, electron, and photon beam–directed nanofabrication and augmentation of materials. An emphasis on understanding the interactions of these energetic beams with 2D and quantum emitters targeted an end goal of enabling precision manufacturing and integration of these materials into functional sensors and elements for next-generation computing. These studies provided critical information about the methods by which these materials may be processed into useful devices using direct-write manufacturing, information which is likely applicable to scalable operations as well. Direct-write nanofabrication as pursued in this project and ongoing efforts to integrate 2D and quantum materials into functional architectures for sensing and computation are relevant to DOE BES's mission objectives as they relate to microelectronics, transformational manufacturing, and quantum materials.

Results and Accomplishments

With respect to electron beam–induced etching and modification of 2D materials, this project looked at 2D systems such as MoS_2 because Mo oxides have relatively high vapor pressure if the team could use

ozone as a precursor for electron beam–induced oxidation of the materials. Upon building and testing a gas injection system for ozone, this study found evidence that the team could oxidize the MoS_2 films, but the by-product volatility was not high enough to etch even with very high beam current (to assist with beam-induced heating and desorption). The team realized that elevated temperature was needed, so the project began using the in situ laser coincident and in parallel with the electron beam–induced oxidation in an attempt to volatilize these oxides. Still, the oxides could not be volatilized even with the addition of photothermal heating.

The team used structures such as localized optical antennae of sorts that could be placed locally near the attempted etch features such that they could absorb the incident light and generate heat in the nanostructure. The quasi-1D nature of the nanostructures limit heat dissipation and become a means to *selectively* photothermally heat a nanosized area relative to the surrounding substrate. However, when the team performed the experiments, they realized that the laser irradiation of the nanostructure was causing what appeared to be a reaction that was localized to the nanostructure surface. This result led to the conclusion that residual precursor deposition gases were spontaneously decomposing on the nanostructures because of their elevated temperature relative to the substrate. To induce selective functionalization of low-dimensional materials such as nanofibers (as were used as a demonstrator), this work successfully pursued a full understanding of the physical constraints of this process for two precursor deposition processes: Pt and Au coating of nanostructures.

Essentially, the team discovered that although this work used electron beam–induced deposition nanofibers as a template for amplifying temperature locally via photothermal heating, a more general phenomena was observed that may be leveraged in low-dimensional materials given they have appropriate optical absorption and have dimensionally constrained heat dissipation limitations. The team performed a thorough investigation of nanostructure geometry and found that the thermal decomposition of organometallic Pt and Au precursors could be accurately predicted based on the absorption and geometric properties of the nanostructure. The team measured the kinetics of the process and found it to be mass transport–limited by the delivery of precursor flux. This work also demonstrated that complex 3D geometries could be fabricated and thermally engineered to selectively drive reactions and specified areas.

This work additionally explored the possibilities of using electron beam–driven chemical etching of 2D materials. The project considered halogenated precursors, beginning with the common Si/SiO_2 etchant gas XeF_2. A key finding of the linearity of etch depth with electron dose is indicative of a relatively stable stoichiometry; one might expect a rate discontinuity if there is a change in the chemistry of the flake. This is a promising result and was further confirmed through Raman analysis, which shows fairly consistent peak intensity ratios with overall intensity scaling with thickness. Because near-atomic layer control of etching was desired in this case, this work attempted to find a precise dose to etch an entire flake and a dose per layer required. Raman and photoluminescence spectra of areas in the flake etched using EBIE indicated a full etch through the entire flake around 5–6 $nC/\mu m^2$.

Some images tended to indicate a nonuniform etching of the flakes, which became increasingly apparent as the etch neared completion. The team hypothesized that even though there was no indication of large stoichiometric changes, the etch mechanism likely involved formation of point defects from which the etch would evolve. As a result, the team exfoliated single-layer flakes onto holey transmission electron microscopy grids and performed a series of etch experiments using a dose below the threshold to etch the complete flake. As expected, the suspended flakes required larger doses to etch than adhered flakes because of the lack of electron scatter in the substrate. Atomic-resolution scanning transmission electron microscopy and image processing confirmed the increase in point defects, likely S vacancies, with dose and an apparent coalescence to larger holes. This result lends credence to the idea that a rate-limiting step may be the beam-induced formation of these point defects, after which the etch rate may accelerate at that locale.

The desirable outcome was not only to etch MoS_2 but also to do so at a high resolution and with limited peripheral damage to the lattice. To test this method, the team fabricated a series of field effect transistors and measured their IV response after the channel had been etched and defined by the EBIE process. EBIE was used to completely remove the MoS_2 between electrodes with varying channel width but equal length. This work found that the electron mobility of the field effect transistor decreased as the width of the channel decreased. Although this result is an undesirable effect, the limit of this peripheral damage owing to electron backscatter is minimal compared with He ion bombardment. Additionally, subsequent simulations found that the electron energy chosen for this experiment was likely far from optimal for minimizing peripheral damage.

11026: Expanding the Cryo-Electron Microscopy Coverage Map By Using Novel Sample Prep and Reconstruction Techniques to Determine the Structural Mechanism of Intracellular Communication

A. Williams

Project Description

The main goal of this project was the development of cryogenic electron microscopy (cryoEM) at ORNL within the Center for Nanophase Materials Sciences (CNMS) to provided state-of-the-art equipment and expertise to ORNL staff and users. The first task of this project was to establish data processing and storage capabilities within CNMS to handle the petabytes of data generated by cryoEM. The second task of this project was to develop modalities of cryoEM (single-particle, tomography, and microcrystal electron diffraction) within ORNL and to establish cross-directorate research interests spanning the material, neutron, and biological sciences. An additional goal of this project was to use single-particle cryoEM to determine the atomic structure of cell signaling proteins and the role that native lipid species play on function. The two signaling cassettes under study were the nuclear receptor LRH1 (liver receptor homologue 1) and its interactions with nuclear phospholipids as well as the cytoplasmic signaling protein MOB1b (Mps one binder kinase activator 1B) with cytoplasmic phospholipids.

Mission Relevance

CryoEM is a leading technique in structural biology for analyzing the conformation of proteins, macromolecules, lipid vesicles, and whole cells in micro- to nanometer ranges. Certain nonbiological systems (e.g., perovskites, Li batteries, zeolites, metal–organic frameworks) are sensitive to electron beam damage and require dose-limited and cryo-temperate data collection. These beam-sensitive materials science samples can benefit from sample preparation, data collection, and data processing techniques developed for biological cryoEM. This project made a cryoEM capability available at ORNL for biological and materials science research endeavors within a DOE user facility.

Results and Accomplishments

This project established safety protocols to work with biological and nanoparticle samples in the cryoEM preparatory lab, the experimental capability, and the data storage and transfer capacity necessary to fully establish the cryoEM-enabled analysis of beam-sensitive materials for DOE user facility users. A paper on the first 3D single-particle reconstruction of polyelectrolytes using cryoEM was published. A paper detailing the 3D structure of cellulose synthase, identified by combining cryoEM with neutron scattering capabilities, was also published.

COMPUTATIONAL SCIENCES AND ENGINEERING DIVISION

10573: High-Performance Computing for Biomedical Health Sciences

A. Kapadia

Project Description

Computational health science is a growing area with a variety of applications. The last few years have seen growth in the influx of human health data from a variety of avenues. Hospital clinical records are generating large multimodal datasets for patients across platforms such as radiological imaging, blood testing, genetic and genomic workup, and patient history at multiple time points. Alongside this data boom, development has occurred in artificial intelligence (AI), machine learning technologies, and graphics processing unit–accelerated computing, which have together led to new methods to analyze and identify unique trends in large datasets. Researchers are at a unique juncture where the confluence of powerful, AI-based technologies, computing power, and large global datasets could yield revolutionary and insightful breakthroughs in human health. With federated and cloud computing becoming more accessible, a comprehensive development of AI methods is needed in healthcare, which includes robust data sources, state-of-the-art AI algorithms, and exascale computational platforms incorporating both federated and high-performance computing architecture for handling protected data. This project focused on three key thrusts for AI methods in biomedical health sciences: (1) AI in medicine, (2) modeling and simulation of biological systems and neutron instrumentation, and (3) synthetic data generation for biomedical research. These thrusts were developed and implemented in a manner that allows expansion to a multitude of application areas with the eventual goal of advancing scalable AI applications in health sciences and high-performance computing research in biomedical and health applications.

Mission Relevance

This project helps address large-scale global health challenges through supercomputing and computational science resources.

Results and Accomplishments

This project developed a computational framework for multiscale modeling of biological and biomedical experiments. The framework combines radiation physics transport simulations in GEANT4 with state-of-the-art anatomic digital phantoms (XCAT phantoms, Duke University). Each phantom contains individual organs and structures defined by a combination of nonuniform rational b-spline (NURBS) surfaces and polygon meshes and can be voxelized at any desired resolution. The phantom anatomy is defined by continuous surfaces, and phantom motion (respiratory and cardiac) is defined by continuous time curves at any desired spatial and temporal resolution. This project implemented the capability for cellular- and subcellular-level radiation dosimetry using computational models in TOPAS-nBio, followed by estimation of DNA repair using MEDRAS. TOPAS-nBio is an extension to GEANT4 that permits modeling of DNA structures, chromatin fibers, and cellular nuclei to evaluate effects of radiation on these structures. MEDRAS is a DNA repair calculator that uses mechanistic models of DNA repair to estimate the postradiation behavior of DNA within specific cell lines. This project integrated COMPUCELL3D to estimate multicellular growth patterns in tumors and tissues in response to radiation damage. Using this multiscale simulation tool, this work generated the first set of radiation dose estimates and DNA damage to specific organs, tissues, and tumors in digital human phantoms following radiation damage from a variety of radiation sources. Estimates were obtained from environmental and atmospheric radiation exposures, medical imaging exposures (computed tomography, x-ray), and targeted radiopharmaceutical treatments using ^{225}Ac (an isotope being developed at ORNL).

CYBER RESILIENCE AND INTELLIGENCE DIVISION

10994: Distributed Internet of Things Systems Key Management

M. Li

Project Description

With the advent of pervasive computing and Internet Protocol version 6, the Internet of Things (IoT) has become an integral part of people's lives as well as numerous business operations. Such a transformation brings on exciting new applications, as well as extra security and privacy concerns. IoT devices typically have limited computation, communication, and storage capabilities. These limitations often lead to security architecture compromises such as using symmetric keys for group membership management. Although secure and efficient in fixed-network settings, state-of-the-art symmetric key solutions, such as the Thread protocol, are ill-adapted to IoT's highly dynamic IoT device membership turnover. Frequent regrouping requires expensive reauthentication, key regeneration, and redistribution to maintain security. Furthermore, whenever IoT members leave a group, the known symmetric keys cannot be forgotten, thus posing a serious vulnerability. As a result, secure and efficient information sharing in such distributed IoT-embedded systems, such as using the publish-and-subscribe framework, becomes a real challenge.

To address such issues, this project proposed Distributed IoT Systems Key Management (DISK), a novel system allowing secure, multileveled IoT group-of-groups key management. DISK is further applied to secure key management in the IoT publisher and subscriber systems. DISK advances the state of the art from local group key management to address the distributed IoT system-of-system secure and efficient key management needs. DISK will advance the security posture of the DOE's IoT-embedded critical infrastructures, such as radioactive/nuclear (R/N) material transportation, by enabling secure and effective information sharing from IoT devices and DOE enterprise servers and human operators.

Mission Relevance

IoT technologies are of paramount importance and interest to DOE operations because IoT permeates multiple interdependent critical infrastructures (oil and gas, communications, water, and transportation). One DOE critical application is R/N material transportation and management, which has emerged as a high-potential candidate and as a major manifestation of the needs of secure IoT technology. In this application context, each R/N package in transport is equipped with edge computing devices and multiple sets of sensors, with secure, real-time network communication and information sharing between the R/N packages' sensor suites and cloud-based (via telematics units) services to support holistic distributed situation awareness in terms of tracking, reporting, integrity validation, and monitoring (e.g., radioactivity, temperature, fire/smoke). Not only can DISK assist DOE R/N material transportation security, but researchers also foresee many other applications, such as securing vehicle ad hoc networking for cooperative navigation and collision avoidance with vehicles coming and leaving frequently. Additionally, this framework could serve in securing patient care networks in modern hospital intensive care unit (ICU) operations, where monitoring/therapeutic IoT devices are continually added to or removed from a group as patients enter and exit the ICU, and their information collected needs to be securely shared with ICU staff.

Results and Accomplishments

The project has completed the design and development a cryptographically based strong key management framework (DISK), allowing for secure distributed IoT group communication and information sharing.

The framework enables flexible IoT group membership management by means of safely adding new IoT devices and securely removing membership of the to-be-deleted devices. Such key management is an improved incremental approach based on Shamir's *K*-out-of-*N* cryptographically strong secret sharing mathematical principle.

The project team has implemented the DISK design in Python. The implementation resulted in a software key management test bed. Researchers have conducted key management validation on this test bed regarding the following capabilities, which are all working correctly.

The initial IoT group construction with a managing node constructed communication secret keys, secret shares splitting, and authenticating and distributing secret shares vectors to initial group member devices. The group is then capable of secure communication via the shared secret key.

The managing node successfully performed new member device additions by the construction of new secret shares vectors and delivering them to the new devices, resulting in new devices effectively and securely joining the IoT group communication.

The managing node, in conjunction with remaining device nodes, successfully performed member device deletion by executing incremental phase shift. The phase shift, coupled with the publication/exposure of the expiring member's secret share, results in insufficient K-out-of-N keys for the expiring member to execute the successful shift to the new phase key required. The revoked member can no longer participate in continual secure group communication and thus is successfully expelled from the IoT group.

The project has further implemented DISK into a publisher-and-subscriber scheme, enabling secure and efficient distributed IoT-embedded system information sharing.

These research results were published in IEEE conference proceedings.[1]

[1] M. Li, S. Hollifield, and M. Iannacone. "Incremental Threshold Scheme Enabled IoT Group Key Management." *Proceedings of the IEEE 7th Cyber Security in Networking Conference*, Montreal, Canada, October 16, 2023.
DOI: 10.1109/CSNet59123.2023.10339701

ENVIRONMENTAL SCIENCES DIVISION

10681: High-Resolution Photosynthesis Data for Improved Bioenergy Assessment

J. Field

Project Description

Bioenergy and bioproducts play a large role in decarbonization scenarios. This role will require growing more woody and herbaceous biomass feedstocks in ways that make efficient use of land, maintain or enhance soil organic carbon (SOC) storage, and limit emissions of N_2O, which is the dominant agricultural greenhouse gas. Process-based agroecosystem models such as DayCent allow the exploration of how energy crop yields, SOC, and N_2O vary in different regions and respond to changes in management (e.g., fertilizer rates, crop rotations). To minimize competition with existing agricultural use, energy crops are targeted toward marginal land. However, land quality is not well-represented in existing models, and observational data are lacking because most field trials are conducted on prime land. This project sought to expand process-based agroecosystem modeling practice and leverage alternative datasets of plant productivity, land biophysical properties, and SOC observations to better assess bioenergy feedstock crop performance and environmental outcomes.

Mission Relevance

Sustainable production of biofuels and bioenergy is integral to the DOE Mission of "ensur[ing] America's security and prosperity by addressing its energy, environmental, and nuclear challenges through transformative science and technology solutions."[1] Cultivation of novel feedstock crops that minimize land use change, such as converting marginal land to perennial biomass crops or cultivating oilseed crops during existing fallow periods in annual crop rotations, is important to goals such as developing sustainable aviation fuel and DOE BETO's aims[2] in quantifying and enhancing SOC storage.

Results and Accomplishments

A capability was developed to use the process-based DayCent model to study and model interactions between plant productivity and SOC storage at landscape scales relative to biofuel and bioenergy production facilities (e.g., *feedstock-sheds*). This capability involves calibrating and validating DayCent based on external observations of energy crop productivity and then using it to predict changes in SOC storage with the cultivation of those energy crops. This modeling approach is broadly consistent with the SOC and greenhouse gas accounting standards used by the Intergovernmental Panel on Climate Change (Tier 3 accounting), the EPA inventory of US greenhouse gas emissions and sinks, and various measurement, reporting, and verification protocols used in voluntary carbon market registries and crediting organizations.

DayCent was used to evaluate the yield, SOC, and N_2O performance of *Brassica carinata*, a nonedible oilseed crop that can be grown as a sustainable aviation fuel feedstock during winter fallow periods in the southeast, and was included in the pending 2023 update of the *Billion Ton Report*.[3] A case study in the tristate area of southern Alabama, southern Georgia, and northern Florida was selected because this region is climatically suitable for existing *B. carinata* varieties, and productivity data were available in this region through partners in the Southeast Partnership for Advanced Renewables from Carinata.[4] This study determined the region can supply approximately 1 billion liters of sustainable aviation fuel annually from *B. carinata*, and climate-smart management could lower its carbon intensity by up to 12 g/MJ CO_2.

[1] DOE. "Mission." Accessed February 19, 2024. https://www.energy.gov/mission.
[2] DOE BETO. "BETO Workshop: Bioenergy's Role in Soil Carbon Storage (virtual)." Held March 28–29, 2022. Accessed February 19, 2024. https://www.energy.gov/eere/bioenergy/events/workshop-bioenergys-role-soil-carbon-storage.
[3] DOE. *2023 Billion-Ton Report*. Draft, 2023.
[4] Southeast Partnership for Advanced Renewables from Carinata. "Home." Accessed February 19, 2024. https://sparc-cap.org/.

DayCent was calibrated based on data from several local field trials and then used to simulate *B. carinata* under different management assumptions across >2 million hectares of cropland in the region, accounting for variability in soil type and weather. After calibrating based on one season of data from a University of Florida agricultural experiment station in Quincy, Florida, the resulting DayCent model was successfully validated against a variety of other *B. carinata* trials (both experimental plots and small commercial plantings) in the region. Associated increases in SOC and changes in N_2O predicted when *B. carinata* is cultivated every third winter instead of leaving the land fallow. This work was published in *Frontiers in Energy Research* in the Bioenergy and Biofuels section.[5]

Additional case studies were conducted for corn ethanol feedstock-sheds around Rensselaer, Indiana, and Craig, Missouri, for evaluating remotely-sensed plant productivity and SOC outcomes.

Three independent, remotely-sensed datasets of landscape-scale plant productivity (Landsat net primary production [NPP], MODIS NPP, and Global OCO-2 based Solar-Induced Fluorescence [GOSIF]) were accessed, analyzed, and visualized in these case study areas via the open-source QGIS modeling program. Ground-based productivity indexes—specifically Land Capability Class and National Commodity Crop Productivity Index (NCCPI)—were also accessed via the NRCS Soil Survey Geographic Database (SSURGO) and included in the comparison. SOC point data were extracted from the World Soil Information Service (WoSIS) Soil Profile Database.[6] Because they are lacking in specific metadata around prior land use history, WoSIS and similar soil profile databases have been underutilized in previous process-based modeling efforts. However, filtering these datasets against current-day land cover and land use maps allows useful inferences to be made. The Cropland Data Layer was used to identify land recently under corn and soy rotations in the case study areas, and mask out all other areas, focusing the analysis on just the most relevant croplands.

The remotely-sensed and ground-based plant productivity datasets examined in this work show minimal consistency with one another. Although the DayCent model-based analysis suggests that SOC changes under bioenergy crops will be strongly influenced by plant productivity, the Rensselaer and Craig case studies suggest that existing plant productivity datasets can be highly divergent from one another, and only certain datasets have predictive value with respect to independent SOC observations. This finding provides important context for ongoing landscape modeling and data assimilation efforts, which have only recently been applied to SOC prediction. The large WoSIS dataset provides independent observations against which to screen the utility of different remotely-sensed data layers for SOC prediction, and perhaps to independently validate process-based model performance. In addition to WoSIS, future work should explore using the NRCS's Rapid Carbon Assessment dataset[7] in a similar capacity. Trials of novel energy crops (e.g., *B. carinata*, switchgrass) are generally conducted at relatively small spatial scales that preclude directly comparison with the remotely sensed productivity datasets listed previously. As these crops become cultivated more widely and at larger spatial scales, it will provide a new opportunity to leverage landscape-scale productivity observations to better quantify carbon cycling, SOC changes, and bioenergy landscape design.

[5] J. L. Field et al. "Modeling Yield, Biogenic Emissions, and Carbon Sequestration in Southeastern Cropping Systems With Winter Carinata." *Frontiers in Energy Research* Bioenergy and Biofuels section 10, 2022. DOI: 10.3389/fenrg.2022.837883
[6]. N. Batjes and L. Calisto. "WoSIS Soil Profile Database." ISRIC World Soil Information. Accessed February 19, 2024. https://www.isric.org/explore/wosis.
[7] NRCS. "Rapid Carbon Assessment (RaCA)." Accessed February 19, 2024. https://www.nrcs.usda.gov/resources/data-and-reports/rapid-carbon-assessment-raca.

MANUFACTURING SCIENCE DIVISION

10686: Efficient Conversion of Carbon Dioxide to Dimethyl Ether

H.-Y. Chen, J. Pihl, T. Toops, S. S. Majumdar

Project Description

Conversion of CO_2 to fuels using renewable H_2 and energy provides an option for the transportation sector to reduce net CO_2 emissions. Dimethyl ether (DME) is a clean fuel and can be used as a substitute for diesel. Direct conversion of CO_2 to DME is thermodynamically favorable for CO_2 conversion and DME selectivity. The direct process requires an active and durable bifunctional catalyst with metal redox components near acid zeolite components. However, under reaction conditions, detrimental interaction between the two catalytic components can lead to rapid catalyst deactivation. The purpose of this project is to design and evaluate layer-structured bifunctional catalysts supported on monolithic substrates. The design involves the coating of the metal redox and the acid zeolite components on the wall of the monolith channels as two distinct layers to minimize the undesirable interaction between the two components. This project demonstrates the concept on commercially viable monolithic substrates. New catalyst compositions and configurations are further optimized as monolith catalysts. An optimized monolith catalyst is expected to exhibit significantly improved CO_2 conversion, DME selectivity, and on-stream durability.

Mission Relevance

This research directly supports the DOE EERE mission to "accelerate the research, development, demonstration, and deployment of technologies and solutions to equitably transition America to net-zero greenhouse gas emission economy-wide by no later than 2050, and ensure the clean energy economy benefits all Americans, creating good paying jobs for the American people—especially workers and communities impacted by the energy transition and those historically underserved by the energy system and overburdened by pollution."[1] It develops an energy-efficient process to convert CO_2 to fuels using renewable H_2 and energy to enable the hard-to-electrify transportation sectors (heavy-duty trucks, off-road equipment, rail, and marine) to meet net-zero CO_2 and near-zero pollutant emissions. The concept can also be applied to convert CO_2 to other sustainable fuels for aviation and marine applications.

Results and Accomplishments

This work developed a concept of layer-structured bifunctional monolith catalysts, in which a $CuO/ZnO/ZrO_2$ component for methanol synthesis using CO_2 as feedstock and a ferrierite zeolite component for the subsequent dehydration reaction are wash-coated on metallic monolith substrates as two separate layers. Such a layered configuration significantly improves the synergistic effects of the two components, resulting in a 20% increase in the production of DME at 240°C compared with the conventional bifunctional catalysts. More remarkably, the layer-structured monolith design minimizes the undesirable interaction between the metal oxides and the zeolite components and drastically improves the on-stream durability of the catalyst. No activity decline was observed during a 146 h performance test. This project further explored the layer-structured multifunctional monolith catalysts for the direct conversion of CO_2 to lighter olefins and other hydrocarbon products. The catalysts exhibit good initial activity but suffer from deactivation, likely because of the formation of coke in the cages of the zeolite components.

[1] DOE EERE. "Mission." DOE. Accessed February 5, 2024. http://www.energy.gov/eere/mission.

10715: Technology Gap Assessment for High-Power Electric Drives in Heavy-Duty Vehicles and All-Battery Electric Locomotives

J. S. Manguelle, B. Ozpineci, V. P. Galigekere

Project Description

This project identified and evaluated the technology gaps for high-power electric drives in medium- and heavy-duty vehicles (MHDVs) and electric locomotives to achieve target specifications and accelerate the adoption of these vehicles. The technological maturity of key electrical components of high-power, commercially available traction drive systems was considered. These components included power electronics systems; propulsion motors, including induction, synchronous, and switched-reluctance types; and sensing and control components. Overall system performance was also considered. This project also assessed the technical needs and proposed technology solutions to increase traction drive power and power density as well as to reduce the charging time of the embedded batteries. A battery charging topology was proposed based on a power conversion building block that can use both wired and wireless power transfer (WPT) interfaces. The following concepts were explored based on modular, scalable, and redundant power conversion architectures: an ultrafast charger for embedded batteries; distributed static WPT; distributed dynamic WPT; grid connectivity, integration, and deployment; and energy management and recovery.

Mission Relevance

MHDVs transport approximately 67% of US freight. They are the second-largest energy use sector, accounting for approximately 23% of total energy use in the United States. In 2019, truck energy consumption was approximately 5,490 TBtu, which was 70% of the total energy use for freight transportation in the United States in 2019. The actual fuel consumption was approximately 25% of total US transportation fuel use during the same year. These numbers show that improving MHDV energy consumption will have a significant positive effect on US energy consumption and emissions. Additionally, according to the Association of American Railroads, railroads move approximately 40% of North America's freight per ton-mile. For each gallon of fuel, railroads can move 1 ton of freight for 470 mi, which is approximately four times more efficient than trucks. However, railroads account for only about 8% of freight-related carbon emissions. Proposing new technologies that increase the electrification of MHDVs and locomotives and improve their energy consumption will tremendously reduce energy consumption in the transportation industry, which will drastically reduce US freight-induced carbon emissions. Primary ways to achieve this goal include developing electric MHDVs technologies, extending the run times of battery-powered locomotives, and increasing the commercial acceptance of these technologies.

Results and Accomplishments

The proposed wireless fast-charging system significantly reduced the charging time of MHDVs and battery-powered locomotives. In this project, distributed static and dynamic WPT power-train configurations were explored. These systems generally had three types of power conversion systems. First was the offboard charging power converter, which converted the grid voltage to a suitable level to charge the onboard energy storage system (e.g., batteries). Second was the direct current (dc)–dc power converter, which converted the battery voltage to a suitable level to supply energy to the traction converter. Third was the traction power converter, which converted existing dc in the vehicle to alternating current with variable frequency and magnitude to drive the electric motors that physically move the vehicle. Because energy recovery was possible in these systems, an energy management, recovery, and balance module was required for system operation.

The distributed static WPT setup was suitable for stationary recharge applications at stations. It featured multiple primary coils strategically placed to transmit power wirelessly to distributed pickup coils and converters. This setup enabled ultrafast charging of battery stacks. The grid interface had a modular architecture with redundancy to enhance reliability. This system is particularly suitable for medium-

voltage power supply applications, providing compatibility with grid infrastructure commonly found in urban and industrial settings.

The distributed dynamic WPT system provided charging capacity while the system was in movement. In this setup, power transmitters were installed along multiple railroad segments, providing power transfer to distributed pickup coils and converters on the moving vehicle. This configuration facilitated ultrafast charging of battery stacks onboard trains or other vehicles, enabling seamless operation without the need for frequent stops for charging. By distributing the receiving coils and converters, this system can ensure uniform charging capabilities across the entire transportation network, enhancing operational efficiency and flexibility. Because of its modularity, scalability, and flexibility, the proposed distributed wireless ultrafast charging architecture can be extended to railroad vehicles by simply stacking more modules in a locomotive. Therefore, a high-power distributed wireless power charging station can be achieved for locomotive applications. A proposed master control strategy was developed to ensure adequate power flow.

The onboard control architecture comprised three control subsystems. The first was the local receiving controller, which was responsible for managing various power electronics converter parameters, such as current or voltage regulation, balance energy transfer, and local pulse-width modulation (PWM) per converter. In addition to the local receiving controller, other local controllers were responsible for modulation and control systems for reversible dc–dc converters, voltage balancing of battery racks, dc–dc PWM interleaving techniques, and traction motor drive control. Lastly, the master system coordination was at the top of the management hierarchy and was responsible for overseeing the operation of the subsystems within the energy network. Its primary functions included coordinating the operation of the local controllers to maintain system-wide stability and efficiency. It also managed system faults by initiating necessary actions such as reconfiguration and adjustment of PWM interleaving to mitigate effects. Additionally, by managing the human–machine interface, the system provided operators with the necessary information and control options for effective monitoring and interaction, thus ensuring smooth system operation and optimized performance.

MATERIALS SCIENCE AND TECHNOLOGY DIVISION

10522: Modeling Evolution Mechanisms in Complex Concentrated Alloys

J. Tiley, S. Nag

Project Description

Refractory complex concentrated alloys (RCCAs) and high-entropy alloys have shown tremendous potential for increasing the high-temperature strength, oxidation resistance, and magnetic properties of structural materials. Researchers are employing diverse material-screening tools to preselect possible material combinations and potential thermomechanical processing procedures. The purpose of this project is to understand and predict the parameters that control phase evolution in chemically complex refractory alloys. The investigation focused on the body-centered cubic (bcc) TiAlNbTaZrHf alloy system with slight elemental variances. This alloy is of interest to researchers because of its potential for stable B2 phases above 1,200°C with high-temperature compression strength and ductility capabilities.

Mission Relevance

The project provided an optimized high-temperature RCCA for consideration in high-strength applications and developed a basic understanding of what underlies the formation of phases and microstructures within RCCAs. New alloys resulting from this research could increase high-temperature strength and damage tolerance capabilities for hypersonic materials, propulsion components, and structural materials. Additionally, the phase evolution and processing knowledge allows for increased optimization of mechanical properties for high-entropy and complex, concentrated materials relevant to DOE's mission.

Results and Accomplishments

The research was centered on phase predictions, evolution, and properties associated with varying Ti compositions. The highest temperature in which the B2 phase was stable occurred with an approximate chemical composition of $Al_{1/3}Ta_{1/3}Ti_{1/3}$. Early CALPHAD modeling results predicted the best composition in the TiAlTaNb system is at approximately 8 atom % Nb and 30.7 atom % of Al, Ti, and Ta each. At this composition, the primary solidified phase is still the bcc phase, and bcc_B2 may form at approximately 1,650°C. Additionally, the best composition in the TiAlTaZr system is at approximately 9 atom % Zr and 30.3 atom % of Al, Ti, and Ta each. At this composition, the primary solidified phase is still the bcc phase, and bcc_B2 may form at approximately 1,700°C.

To better understand the effect of the Al and Ti ratio on phase formation, ternary TiAlTa samples were created with differing Ti composition (while keeping Al and Ta ratios constant). The homogenized microstructure of the 33Ti shows large, blocky precipitates (phase 1) with intermittent cracks within the gray matrix (phase 2). As the composition shifts to 50Ti, three different phases are visible: (1) Ti-rich, stubby precipitates that show light gray contrast (phase 1); (2) high, Al-containing, dark gray precipitates (phase 2) that have a relatively sharp and corrugated interface; and (3) a brighter, Ta-rich matrix.

Microstructure evolution was explored through scanning electron microscopy, x-ray diffraction, and transmission electron microscopy characterization. Several ordered intermetallic phases were identified in the TiAlTa system, consistent with existing literature. Notably, eutectic-type microstructures were observed in the 33Ti samples, possibly influenced by cooling rates during fabrication. The top edge of L33Ti, which was the last to solidify during melting, and the s33Ti sample from the SA material core, which was also the last to solidify during processing, did not have these structures, indicating a possible kinetic relationship. The presence of eutectic-type microstructures may have contributed to reduced oxidation resistance, as indicated by the team's oxidation results.

Subsequent nanoindentation experiments provided valuable insights into the nanoscale mechanical properties of the materials at elevated temperatures. The s33Ti and s70Ti displayed the highest Young's modulus and the highest hardness value, respectively, at room temperature. At higher temperature, oxidation generally increased the hardness of the materials with the exception of s70Ti, where lower oxidation did not offset temperature-dependent strength loss. Interestingly, the Ta_2Al phase exhibited a nanoindentation hardness value nearly twice that of the TiAl phase at room temperature.

This investigation delved into oxidation behavior with weight-gain analysis and x-ray photoelectron spectroscopy analysis shedding light on oxidation resistance. Among the alloy variants, L70Ti exhibited the highest oxidation resistance, and L50Ti samples displayed the lowest resistance possibly because of the preferential generation of Al_2O_3 over TiO_2. Interestingly, the s33Ti alloy provided higher oxidation resistance compared with other L33Ti samples. However, notably, sample preparation may have affected these results.

Characterization of oxidation phases, including x-ray photoelectron spectroscopy and scanning electron microscopy–energy dispersive x-ray spectroscopy analysis, provided complementary insights into the oxidation behavior of TiAlTa alloys. This study found that the segregation of Ta oxides could lead to larger cracking and create a pathway for oxygen penetration into the L33Ti alloy. Cooling rates during the oxidation tests and during initial synthesis of materials affected both microstructure and oxidation behavior. Typically, allowing samples to furnace-cool instead of air-cool before weighing increased their oxidation rates and resulted in material spalling from several samples. However, furnace-cooled L70Ti had higher oxidation rates than the air-cooled sample possibly because of the increased time within the furnace. This result has implications on future applications involving thermal shock conditions.

In summary, Ti additions and modified cooling conditions significantly affected the kinetics of both oxidation behavior and phase evolution in TiAlTa alloys. The optimization of the resulting phase evolution may provide critical high-temperature bcc and B2 phases that enable increased strength and oxidation resistance at high temperatures. The findings emphasize the importance of considering composition, microstructure, and sample preparation techniques in optimizing alloys' mechanical and oxidation properties. Further research is necessary to enhance the accuracy of thermodynamic modeling and to uncover the underlying mechanisms associated with the observed evolution phenomena.

10552: Tuning Quantum Magnets By Controlling Disorder

J. S. Gardner, V. Cooper, J. Paddison

Project Description

The overarching goal of this project was to understand how atomic disorder controls emergent quantum properties of matter, including the magnetic monopole and skyrmion motion. Of particular interest here were geometrically frustrated triangle, kagome, and pyrochlore magnets in which the perfect lattice promotes a highly degenerate spin liquid ground state when the sublattice is decorated with antiferromagnetically coupled spins. Along with chemical substitution to control disorder, samples were synthesized in various atmospheres up to 150 bar to tune the oxygen sublattice. Polycrystalline and single-crystal samples were synthesized and characterized, and then studies at the atomic level were performed at neutron, muon, and synchrotron x-ray sources.

Mission Relevance

This project aligns with objectives outlined by DOE BES in its 2018 Roundtable on Opportunities for Quantum Computing in Chemical and Materials Sciences. Using probes (e.g., neutron scattering, muons), this work proposed to understand how this disorder controls emergent quantum properties of matter, including the magnetic monopole and skyrmion motion. This work developed low-temperature characterization tools to probe the bulk magnetic properties of the samples made and models to characterize the disorder in powders and crystals at the 2% to 3% level. As such, the tools developed are

also relevant to the radiation damage community, the electrolyte community, and those using scattering techniques to study short-range correlations.

Results and Accomplishments

This project grew and characterized various crystalline materials. Within these materials, this work attempted to control and measure disorder. The materials were characterized with lab-based x-rays, the temperature dependence of the bulk magnetization, dynamic susceptibility, and specific heat. This project performed experiments on the Low-Energy Muon spectrometer and was able to observe the depth dependence to magnetism in $PdCoO_2$ films. These data complemented neutron reflection results but also allowed the team to nondestructively probe the internal spin dynamics. Here, the stoichiometric parent compound was nonmagnetic, and defects to the crystalline lattice induced bulk magnetism. In another system, the model magnet $Gd_2Ti_2O_7$ was used to model the magnetic sublattice at a variety of length scales. This project was able to correctly fit the neutron diffraction data only with an understanding of local and long-range correlations.

10700: Hybrid Microstructural Evolution for Site-Specific Property Response

S. Nag, B. Jordan, K. An, J. Haley, Y. Lee, J. Tiley

Project Description

This research investigated a combinatorial approach of zone-based hybrid microstructural evolution using a directed energy deposition additive manufacturing (AM) technique. This technique is a truly material-agnostic concept and may be employed to fabricate graded, high-temperature metallic and intermetallic parts comprising high-strength materials, including Ti alloys, Ni superalloys, and Nb- and Mo-based refractory alloys.

Mission Relevance

Currently, going from part concept to production of mission-critical additively manufactured components can take several years. This time length is prohibitive to the broad adoption of AM technologies for use in sustainment applications, where cost and readiness are vital to compete with conventional processes. To make AM a substantial manufacturing strategy, researchers must incorporate its unique attributes of optimizing materials (composition), design, and manufacturing modalities to fit site-specific performance needs. Keeping these aspects in mind, this project aimed to investigate additively built blocks with site-specific compositional gradation. The project had a broader vision to integrate breakthrough AM functionalities for a wide range of applications in domains such as energy (e.g., marine, nuclear, and renewables), space (e.g., access, high velocity), and aviation (e.g., structural engine components).

Results and Accomplishments

Multimodal in situ sensing was improved for directed energy deposition AM of graded materials. The existing in situ sensing on the AM system used to measure component temperature, melt pool characteristics, and external strain fields through digital image correlation; this system was improved to enable multimodal sensor fusion. These in situ data enable faster parameter optimization through the direct measurement of relevant physical phenomena that lead to failure. This capability greatly enhances the capacity for AM materials exploration by shrinking the time required for development cycles.

11006: Environmental Degradation of Structural Materials: Exploring Nanoscale Reactions in Liquids and Gases Using Advanced In Situ Analytical Electron Microscopy

G. Burke, Y.-R. Lin, K. Unocic, T. S. Byun

Project Description

This project established and demonstrated a robust in situ liquid methodology and platform for structural materials research involving the initiation of corrosion, oxidation, electrochemical polarization, and H effects that can be applied to a variety of technologically important structural materials to develop fundamental understanding of environmental and material reactions under conditions. In this project, some specific alloys (e.g., Al–Si, austenitic stainless steels, low alloy steels) were selected to examine considering material needs for low-C energy and H-related applications. This effort particularly focused on demonstrating the ability of liquid in situ and gas-phase in situ analytical electron microscopy to provide unique information on the nanoscale reactions (precursor reactions) leading to the initiation of material degradation. The in situ experiments were complemented by analysis of bulk test specimens to ensure that the observations were pertinent and applicable to the degradation phenomena under investigation.

Mission Relevance

This project is directly relevant to advanced reactor materials research, fossil energy materials research, and materials research associated with the DOE Hydrogen Shot (materials for the H economy). This project addressed the experimental hurdles in the study of the precursor reactions associated with the initiation of materials degradation in liquid and gaseous environments and aimed to assist in the ultimate development of models to address the initiation of degradation and environmentally assisted cracking in real structural materials. It facilitated further research with application-specific materials for degradation in (1) existing and advanced nuclear power systems; (2) H-containing environments associated with H cogeneration, H transport, and gaseous H storage; and (3) fossil energy systems (H/natural gas line pipe steels and welds).

Results and Accomplishments

This project installed the world's first Protochips Poseidon Select Liquid Electrochemical Cell with an electron dose monitor system for in situ transmission electron microscopy (TEM) experiments at the Low Activation Materials Development and Analysis Laboratory at ORNL. Studying materials degradation in liquid cell TEM requires meticulous isolation of material behavior from electron beam effects, such as beam heating and radiolysis. The high-intensity electron beam can induce various interactions with the sample, leading to contamination, bubble formation, and sputtering. To address this issue, a TEM holder with a Faraday cup was used to measure and calibrate the electron beam current under different imaging conditions. Accurate beam current calibrations linked to AXON software ensured optimal accuracy and precision for dose quantification during in situ TEM experiments. The AXON software provided live updates of the total accumulated electron dose and electron beam–exposed area during imaging. Additionally, the electrical performance of the system was assessed through single-frequency electrochemical impedance spectroscopy and open-circuit potential testing, confirming system stabilization and electrical behavior of the solution at the sample. Careful initial calibration and testing were crucial to minimize and detect artifacts in liquid electrochemical cell TEM experiments. Establishing a robust in situ capability necessitates the development of appropriate experimental protocols and benchmarking research, which includes understanding the effects of the electron beam on the system.

When preparing TEM samples using focused ion beam (FIB) methods, damage caused by Ga ions can interfere with the liquid electrochemical reaction and obscure the desired features. Therefore, ensuring artifact-free specimen preparation is crucial for conducting in situ liquid electrochemical cell experiments. As part of the methodology development in the project, this work developed a flash electropolishing

method and procedure to effectively remove or minimize FIB damage layers from the surface of FIB-TEM samples. This technique has been successfully implemented across various materials, including Fe, Fe-based model alloys, commercial Fe–Cr alloys, advanced Fe–Cr alloys, and stainless steels. Furthermore, this project investigated the application of scanning TEM–weak-beam dark-field methods. These methods have proven effective in minimizing background contrasts, isolating defect information for dislocation loop classification, providing detailed dislocation line images for small loop analysis, and offering inside–outside contrast for loop characterization. The advancement of flash electropolishing TEM specimen preparation techniques and weak-beam dark-field–scanning TEM imaging methods extends beyond in situ liquid cell TEM experiments, offering significant benefits for microstructure characterization studies.

Ex situ experiments, conducted with the electron beam off to eliminate beam-related effects such as beam heating and water radiolysis, have been performed using the versatile Protochips Poseidon Select Liquid Cell. These experiments involved stainless steel samples, both with and without MnS inclusions, immersed in deionized water. The open-circuit potential tests conducted on the stainless steel samples clearly revealed a distinction between those with and without MnS inclusions when subjected to flowing liquid over time. This disparity can likely be attributed to the dissolution of MnS in deionized water. These ex situ experiments serve as benchmark studies, confirming whether additional changes can be observed in in situ TEM experiments when both the samples and the liquid cell are exposed to the electron beam.

This project successfully carried out a series of liquid cell TEM experiments aimed at real-time observation of Ag crystal growth under liquid flow. These experiments involved simultaneously recording the electron beam dose and open-circuit potential as a function of time. The initial liquid cell TEM experiments focused on Ag crystal growth, serving as a critical step in validating parameters such as liquid flow rate, as well as the size of inlet and outlet tubes to ensure that the cell remained intact within the microscope under liquid pressure. Future plans for this work involve the preparation of stainless steel samples, both with and without MnS inclusions, using flash electron polishing methods to minimize sample artifacts. Subsequently, the team plans to conduct in situ liquid cell TEM experiments and compare the results with those obtained from ex situ experiments.

NEUTRON TECHNOLOGIES DIVISION

10629: Novel High-Rate Neutron Beam Monitor

Y. Diawara, V. Sedov, A. Trofimov, J. Beal, L. Funk

Project Description

This project described two concepts of a novel high-rate neutron beam monitoring system exceeding 1,015 counts per second with subnanosecond time resolution. This project consisted of using optimal neutron reactive layers to convert neutrons into charged particles. The detector may be used to detect either thermal neutrons via nuclear reactions or fast neutrons. The first design was based on a Schottky, homo, or hetero junction type semiconductor device. The neutron reactive layer (made of Li or B) was incorporated in the entrance window of such a charged particle detector and acted as the neutron convertor. The compactness and light weight of such a detection system are attractive characteristics for a handheld, pocket-sized device (i.e., miniature), but the system is scalable to reach larger sizes. In another embodiment, neutron-reactive layers (such as a degenerated P^+ crystalline Si) served as a cathode in two parallel plates in an avalanche chamber design, in which occurred the release of primary charged particles. These charged particles were converted into primary electrons in the gas and amplified by an internal gas multiplication effect to register individual neutrons. Both options were designed to obtain subnanosecond charge transit times, and the signal registration was performed with a preamplifier, a shaper, and a dual-threshold constant fraction discriminator followed by a baseline restorer, counter, and a time-stamper. This beam monitoring system has applications in neutron scattering facilities or in security for dose measurement in counting mode from a single event up to the highest nuclear accident or blast.

Mission Relevance

This project addressed the need for a high-rate beam monitor operating in transmission mode with low efficiency (10^{-2} to 10^{-8}). At neutron scattering facilities, neutron beam monitors are essential for scattering data normalization, beam diagnostics, commissioning, and many other applications. This high-speed neutron detector technology benefits current Spallation Neutron Source and future Second Target Station beamlines, and the technology could be transferred to industry for commercialization. After successful completion of this prototype, the high counting rate and high-resolution features will be most valuable for time-resolved experiments in neutron scattering applications. Beyond these applications, this detector offers three other unique features: (1) a large and high-rate beam monitor using segmented (quadrants) option for beam profiling, (2) a compact and lightweight beam monitor that can be used as a dosimeter (medical applications), and (3) a replacement for ^{3}He by a solid converter, which fills a growing need for neutron scattering facilities worldwide and expands the application beyond neutron scattering to many other fields (homeland security, industrial, medical, and more).

Results and Accomplishments

The tests on the high rate capability were successfully demonstrated, and the imaging feature needs to be tested to fully complete the project.

Many proof-of-principle beam monitor prototypes were fabricated consisting of 10 × 10 mm Si-based photodiodes, on top of which thin neutron reactive layers were deposited. Various layers have been tested (^{10}B, ^{6}Li, natural B) to meet the desired efficiencies.

The time-of-flight spectrum of a ^{3}He-based neutron beam monitor was compared with Si-based and ^{6}LiF-coated on Si–based neutron beam monitors monitor obtained at a Spallation Neutron Source beamline (BL-3). The expected positions of the 220 and 211 Al absorption edges were shown in the spectrums obtained in this work. The Si-based monitor showed a gamma peak in the 0–0.5 Å range, which was absent from the ^{3}He monitor spectrum.

Additionally, three prototypes of 100 × 100 mm Si-based neutron beam monitors were prepared. They are double-sided Si strip detectors with thin ^{10}B, ^{6}Li, and natural B layers coated on one side. The electronic readout was integrated into the detectors for testing.

RESEARCH ACCELERATOR DIVISION

10653: Advanced Concepts and Technologies of Superconducting Radio Frequency Proton Linear Accelerators

R.-L. Geng

Project Description

This project aimed to develop advanced accelerator concepts for the next-generation superconducting radio frequency (SRF) proton linear accelerators (i.e., linacs), enabling their applications to provide societal benefits in areas such as medical treatment and accelerator-driven systems. These concepts were based on the successful operational experience of Spallation Neutron Source (SNS) SRF linacs. Therefore, the project positions ORNL to lead the effort in addressing important societal needs by repurposing SRF linac capabilities and expertise developed originally for basic research. The accelerator design requirements in reliability and availability for societal applications are much more stringent. Meeting those requirements, especially for accelerator-driven system–class, high-power (10–20 MW) proton linacs, is an outstanding challenge. SRF technology is regarded as the most promising path forward. This work identified problem areas that require new technologies, developed new concepts for highly reliable SRF systems, and suggested directions for future development activities. The successful completion of the described work led to the demonstration of an advanced, low-energy SRF proton linac, immediately relevant to medical application areas, including the next-generation proton beam therapy and accelerator-based production of medical isotopes.

Mission Relevance

This project could ultimately deliver a wide range of societal benefits relevant to DOE's mission to ensure America's security and prosperity by addressing its energy, environmental, and nuclear challenges, from medical applications to nuclear waste reduction and, perhaps, even to energy production.

Results and Accomplishments

The SNS SRF linac reliability and availability model was improved. The mean time between failure of the SNS superconducting linac was obtained for the 12-year period from FY 2011 to FY 2022. The project continued the collaborative effort to improve consistency between the superconducting linac and the normal conducting radio frequency linac systems. The analysis on the downtime data derived from the machine protection system was initiated. The team visited Jefferson Lab to participate in the first experimental test of the advanced liquid N cleaning concept. The first trial of depositing Cu alloy onto a thin sheet of Nb was conducted at ORNL's Manufacturing Demonstration Facility using an additive manufacturing method. A strong bonding between the deposited Cu alloy and the substrate Nb was demonstrated by a bending test and a liquid N cold shocking test.

10908: Feasibility Study of High-Power Hadron Accelerator

V. Morozov, A. Aleksandrov, S.-H. Kim, S. Cousineau, A. Shishlo, C. Peters, R.-L. Geng, K. Ruisard, F. Lin

Project Description

The feasibility of a novel, compact, high-power linear proton accelerator (i.e., linac) with an average power of 10 MW at a kinetic beam energy of 1 GeV was explored in this project. Acceleration was accomplished continuously in several stages. In each stage, the beam was recirculated several times through the same set of superconducting radio frequency (SRF) cavities until the cavities' available radio frequency (RF) power was fully used. Depending on the accelerating stage and acceleration efficiency, the number of recirculations could be quite large, making individual recirculation arcs impractical. Therefore, this project adopted the fixed-field alternating gradient (FFA) arc design, in which all the

passes within a large, continuous momentum range could be transported by a single beamline. Similarly, the conventional approach of connecting an arc to a linac section by separating different passes into individual beamlines became impractically complex. Instead, this work implemented an adiabatic matching technique whereby the transfer of all beam passes occurred simultaneously within a single beamline. This project introduced a novel concept for control of the orbit and optics of individual passes by their harmonic excitation. This study outlined approaches to optimization of linac timing and control of the longitudinal beam dynamics.

Mission Relevance

The development of a 10 MW facility could be the next logical step after the completion of the Proton Power Upgrade program at the Spallation Neutron Source. It is particularly interesting from a practical point of view because it would enable accelerator-driven subcritical reactor applications. This work could provide solutions to clean energy generation, disposal of spent nuclear waste, and production of liquid fuels. One way to reach this goal in a compact and efficient way is to use a multistage proton recirculating linear accelerator. This accelerator allows for the reuse of SRF cavities, which are one of the most expensive components of an accelerator design. The complexity of multiple recirculating arcs for the individual passes required in a conventional design was solved by confining all passes to a single beam pipe using the FFA concept.

Results and Accomplishments

This project developed the concept of a new kind of high-power hadron linac in which the cavities in different sections of the linac operate near their power and, therefore, current limits, thus fully using the available RF power. The linac was divided into several sections, and each section was individually optimized by recirculating the beam through its cavities several times until the combined current of all passes approached the current limit of the section's cavities. At the same time, the number of cavities in each section was reduced compared with the single-pass case by a factor of one over the number of passes. Beam recirculation significantly reduced the required number of cavities compared with a straight linac case and therefore made the footprint and cost of the entire facility much smaller.

The maximum number of recirculating passes through a particular linac section and, therefore, its maximum attainable energy gain is limited by several factors. This project mitigated the problem of having to deal with multiple return arcs by applying the FFA approach to the arc design. This approach allowed for the transport of multiple different-energy beams by a single beamline. An FFA arc has a wide momentum acceptance with the maximum-to-minimum momentum ratio of over a factor of two. This way, multiple individual return arcs were replaced by a single arc carrying multiple beam passes at the same time.

A conventional approach of matching an FFA arc to a linac involves separating different-energy passes into individual channels and adjusting the optics of each pass independently. This adjustment is accomplished in the recombiner and spreader sections at the entrance and exit ends of the linac, respectively. However, with a large number of recirculations, the spreader and recombiner designs became impractically complex. The challenge using the FFA principle to match an FFA arc to a linac in a single beamline is that all beams must be controlled simultaneously by common magnets. This project solved this issue using an adiabatic matching method to gradually transform between the arc and linac optics over a continuous momentum range. Thus, the combination of an FFA arc and such a transition section supported any number of passes within the FFA's momentum acceptance. Fine-tuning of the parameters of each pass was implemented using harmonic correction. A sequence of correctors was powered in such a way that their kicks were synchronized with free betatron oscillations of a particular pass. With a sufficiently large number of correctors, this process allowed for selective control of a single pass without perturbing the others. This design also provided the flexibility in solving the next issue—namely, tuning the linac timing.

In a straight linac, the RF cavity phases are tuned to provide continuous beam acceleration, accounting for the change in the beam velocity along the linac. In the recirculating scenario, the initial beam velocity changes sufficiently from pass to pass such that it is not possible to tune the linac for maximum acceleration of all passes simultaneously. This project overcame this problem by tuning the linac in such a way that the net acceleration goal is reached by deviating from the maximum acceleration setup for each pass but, at the same time, increasing the number of passes to reach the next acceleration goal. Such flexibility in the number of passes was provided by the FFA return arc design. Additionally, the freedom in the choice of the number of passes, initial timing of each pass, and intercavity timing allowed the project to satisfy the final major constraint—namely, the cavity input power limit. This work explored several approaches to select optimal linac timing, including serpentine acceleration at low energies, harmonic jump at high energies, analytic considerations, genetic algorithm optimization, and splitting each linac into several parallel digital-momentum sections, each covering its own part of the overall momentum range.

Finally, industrial, medical, and accelerator-driven subcritical reactor applications of high-power proton linacs require high reliability and availability of the accelerator beam. For example, for industrial-scale power generation, the number of beam trips for longer than 5 min should not exceed three trips per year. With sufficient energy margin built in, the flexibility of the FFA-based recirculating acceleration scheme in terms of the number of passes in each RLA stage, energy gain per pass, and linac timing should allow for a quick reconfiguration of the accelerator setup to account for and eliminate a single point of failure such as a failed SRF cavity. This mitigation can reduce the duration of beam interruption periods.

10909: Energy Recovery Linear Accelerator-Based Compact X-Ray Free-Electron Laser

F. Lin, A. Aleksandrov, S.-H. Kim, S. Cousineau, A. Shishlo, V. Morozov

Project Description

A free-electron laser (FEL) is a tunable source of coherent radiation that uses a bunch of relativistic electrons to resonantly amplify an electromagnetic wave through magnetic field undulators. The FEL holds great potential for applications as a tunable, high-power, coherent source because of its short-wavelength radiation. It extends the limitation of fully coherent laser light sources in the wavelength of infrared, visible, and near-ultraviolet ranges to the x-ray range with a stable and well-characterized temporal structure in the femtosecond time domain. In particular, the x-ray laser, with a femtosecond pulse length and coherent photons at the angstrom wavelength, opens a new window on the exploration of matter at a length scale corresponding to the atom size and with the dynamics of atomic and molecular processes on their own time scales in many scientific fields.

The x-ray FEL (XFEL) process strongly depends on the local electron beam properties: current, energy, emittance, and energy spread. All existing XFELs are driven by linear accelerators for the purpose of preserving electron beam qualities to achieve a high peak brightness. Normal conducting radio frequency (RF) cavities, with very high accelerating gradients of up to 60 MV/m, are used to keep the linac length as short as possible. However, the normal conducting RF cavities limit the bunch repetition rate up to about 100 Hz in a pulsed beam operation mode, resulting in an average photon brightness of as much as 10 orders of magnitude lower than the peak one. Additionally, the electron beam average current is limited by the klystron input power and beam dump power (practically less than 1 MW) in a linac. Thus, high-gradient superconducting RF (SRF) technology is considered actively in the FEL community to achieve a continuous-wave beam operation mode, which consequently increases the average photon brightness by several orders of magnitude while keeping the same or higher peak brightness. Another significant advantage of using SRF technology is its unique capability of energy recovery, with the beam energy being transferred back to SRF cavities for the acceleration of subsequent low-energy beams.

Therefore, this project proposed an energy recovery linac (ERL)-based XFEL facility. The advantages of this concept can be summarized as (1) shortening the linac by recirculating the electron beam through high-gradient SRF cavities, (2) saving the klystron power and reducing the beam dump power through the energy recovery in SRF cavities, and (3) producing a high average photon brightness by a high average beam power. The most novel of these concepts is its capability of having simultaneous, multiple, continuous-wave XFEL sources with optimized high-brightness performance at a particularly low power consumption. The goal of this project is to study the feasibility of such a concept in the following aspects: optics design, parameter optimization, beam dynamics study, R&D identification, and performance evaluation.

Mission Relevance

The high-gain FEL has been developed as a new generation of an x-ray facility. Its extremely high brightness and well-established coherent photon beam reaches to the angstrom level of a wavelength with a femtosecond-level pulse length, allowing observations of matter on the spatial and time scales of atomic and molecular processes. To generate the desired photon light, the requirements on the electron beam are rigorous in beam size and energy spread. All operating XFELs are based on a linac design to minimize the quality degradation of electron beams. This design results in an accelerator facility large in size, inefficient in energy budget, and with unoptimized performance. This project proposed the development of an ERL-based compact XFEL. Success in this project provides the possibility of building an x-ray photon source with ultrashort wavelengths and high peak and average brightness. This outcome will benefit the communities of scientific users in materials science and many other research fields.

Results and Accomplishments

The injection optics for a 250 MeV electron beam energy was developed to minimize the beam quality degradation owing to either the space charge effect or the optical emittance growth (or both) and to provide the first electron bunch compression. The required dipole and quadrupole fields in this relatively low injection beam energy are small, with a maximum dipole field of 0.064 T and maximum quadrupole field of 1.8 T/m. The optics has $M_{56} = 0.239$ cm that can compress the bunch by a factor of six with the help of a 3.9 GHz chirping cavity running at 12.8 MV. Sextupoles are placed in the large horizontal dispersion regions to adjust the particle distribution in the longitudinal plane because of the effect of second order dispersion T_{566}.The root mean square bunch length was compressed from an initial 600 μm to 95 μm after the chicane, and the root mean square energy spread increases from 4×10^{-4} to 25×10^{-4}. The energy spread will be naturally damped to 2.8×10^{-4} when the beam is accelerated to 2.25 GeV after the first acceleration pass in the ERL.

The linac optics design for both acceleration and deceleration of an electron beam from 250 MeV to 6.25 GeV, and vice versa, was completed. Constraints were applied in the process of design and optimization to control the beta functions in the whole energy range, including (1) fixed quadrupole magnetic fields, (2) a symmetric quadrupole arrangement with respect to the center of the linac, (3) a practical cavity energy gain/loss of 16 MV/m, and (4) one quadrupole between two adjacent cavities. With the maximum quadrupole field gradient of 2 T/m, the beta functions in the linac can be greatly controlled under 100 m in the whole energy range.

The optics of the whole accelerator complex were accomplished by connecting three major sections (i.e., injection, linac, and arcs). Front-to-end particle tracking simulations in this accelerator system are carried out considering both incoherent and coherent synchrotron radiation effects. Degradation of beam qualities, emittance, and energy spread is moderate because of the incoherent synchrotron radiation effect; however, this is rigorous because of the coherent synchrotron radiation effect. Therefore, the most potential R&D aspect is further optimization of design in arc optics on controlling emittance and energy spread degradation owing to the coherent synchrotron radiation effect, linac optics on providing a large energy range of acceleration and deceleration, and injection optics on potential improvement of injecting and merging beams with a substantial difference of beam energies.

A bunch splitting and recombining structure for RF separators and recombiners has been developed. A minimum of three RF frequencies are sufficient to produce two XFEL photon sources at every desired beam energy of 2.25, 4.25, and 6.25 GeV. The required RF frequencies are 1.5 GHz, 750 MHz, and 500 MHz. Technology on producing and manufacturing SRF cavities with these three frequencies is mature, and such cavities are in operation in many accelerator facilities. SRF cavity-related challenges on beam-breakup and high-order mode are evaluated and compared with past, current, and future facilities. The dipole and monopole impedance budgets in this proposed ERL XFEL facility are in between two past successfully operated accelerators—the Jefferson Lab FEL and 6 GeV CEBAF era.

SUMMARIES OF PROJECTS SUPPORTED BY DISTINGUISHED STAFF FELLOWSHIPS

RUSSELL FELLOWSHIP

10906: Evolution of Abiotic Stress Tolerance in Plants

B. Piatkowski

Project Description

Advances in molecular and computational biology have provided powerful techniques to identify the genes associated with traits of interest; however, integration of such findings across different organisms remains difficult because of the complex genomic background unique to each species. The use of evolutionary methods can help to project gene-to-trait associations across levels of biological organization and gain a predictive understanding of how such associations are modified by the environment or by interaction with other organisms. Among the most important traits to the DOE BER program is plant response to abiotic stress because of its relevance to the sustainable production of bioenergy feedstock. The purpose of this project was to apply evolutionary techniques to better understand how plants respond to challenging environmental conditions and to identify the genetic components of abiotic stress tolerance that are shared across levels of biological hierarchy. The goals were to determine the core gene modules responsible for abiotic stress response across plant lineages, elucidate how symbiotic microbes reprogram those modules to confer stress tolerance, and describe how natural selection acts to shape module expression in plant populations along environmental gradients. This project has provided a framework to map gene-to-trait associations across a phylogeny, to better understand the interaction between plants and their environment, and to address how complexity emerges across biological scales.

Mission Relevance

This project aligns closely with the DOE BER program missions of providing sustainable feedstocks to produce bioenergy and obtaining a predictive understanding of how plants interact with their environments. A major contribution of this research is that it enables the translation of functional genomic discoveries across model systems and their projection into novel systems. Furthermore, insights from this research into how microbes influence plant gene expression and how natural selection shapes gene regulatory networks during the process of speciation will help tenable bioengineering of crops and feedstock plants to increase their tolerance to stress in changing environments.

Results and Accomplishments

Data analysis was completed for the core abiotic stress gene modules in plants. The results were published in a journal article.[1] Work regarding the effect of bacteria on plant thermotolerance has also been published.[2] Genome sequencing data were used to demonstrate adaptive differentiation and speciation in plant populations as a response to climate. A new methodology and code were developed to (1) compare gene expression profiles across divergent plant species, (2) model how microbial inoculation affects plant host gene expression, and (3) elucidate genomic patterns of climate adaptation in plant populations.

[1] R. B. Davidson et al. “Predicted Structural Proteome of *Sphagnum divinum* and Proteome-Scale Annotation.” *Bioinformatics* 39, 8, 2023, btad511. DOI: 10.1093/bioinformatics/btad511

[2] B. Piatkowski et al. “Divergent Selection and Climate Adaptation Fuel Genomic Differentiation between Sister Species of Sphagnum (Peat Moss).” *Annals of Botany* 132, 3, 2023, 499–512. DOI: 10.1093/aob/mcad104

WEINBERG FELLOWSHIP

10441: Improving Nuclear Materials Development Cycle with High Throughput Microscopy and Machine Learning

S. Taller

Project Description

The amount of time required to understand life-limiting degradation mechanisms for advanced nuclear reactors must be reduced. Many of the proposed advanced nuclear power reactor designs depend on structural materials to withstand high temperatures and intense radiation fields. Therefore, the grand challenge is to understand the mechanisms of radiation-induced changes in materials in order to assess and predict material performance in these proposed designs. The goal of this project is to advance nuclear technology via accelerated development of radiation-resistant materials. To accomplish this task, a combination of emerging and established technologies must be used to evaluate materials rapidly. This project includes an investigation of the stability of precipitates in additively manufactured Ni–Fe–Cr alloys and their use as trapping sites to mitigate He embrittlement. This investigation is conducted using thermal aging and irradiation followed by postirradiation examination enhanced with machine learning (ML). This project develops tools for automated electron microscopy and defect identification powered by artificial intelligence (AI). These tools elucidate the processes governing He embrittlement in a fraction of the traditional time required to investigate radiation effects.

Mission Relevance

The mission of the DOE is to ensure US security and prosperity by addressing its energy, environmental, and nuclear challenges through transformative science and technology solutions. To fulfill this goal, ORNL set its mission to deliver scientific discoveries and technical breakthroughs that will accelerate the development and deployment of solutions in clean energy and global security as well as create economic opportunities for the nation. The first part of the project addresses the targeted research directions of the discovery and design of new materials for energy applications and the initiative to accelerate R&D and manufacturing for energy systems. The poor machinability and extensive work-hardening of Ni alloys makes advanced manufacturing an attractive option for producing geometrically complex components. Advanced manufacturing of Ni alloys is anticipated to reduce the necessary processing steps for application-specific components. The second part of the project addresses the need for breakthrough nuclear technologies by using ML to accelerate postirradiation examination. Using advanced manufacturing and AI together matches this work with the initiative to accelerate R&D and manufacturing of energy systems.

Results and Accomplishments

The original scientific and technical hypotheses of this project were that (1) radiation damage will enhance the nucleation of precipitates from the solid-solution alloy and dissolve existing precipitates in those already containing precipitates; (2) because the sequestration of He is proportional to the surface area of the precipitate–lattice interface, precipitate dissolution will enhance He embrittlement; and (3) a trained AI will identify defects with the same accuracy as human experts in a fraction of the time.

The first hypothesis was evaluated through the comparison of the homogenized alloy in its as–heat treated, aged, and irradiated conditions. The advanced manufactured superalloy 718 was treated to have a high density of γ″ precipitates to strengthen it or homogenized to have no strengthening phases present. Aging did not significantly alter the properties of the precipitate-strengthened heat treatment, nor did high-temperature irradiation. In the initial homogenized state, low-temperature aging did not alter the properties. However, aging and irradiation near 600°C induced γ″ precipitates and increased the strength by approximately a factor of two, resulting in similar mechanical performance. However, aging-induced γ″ precipitates consisted of a high density of small precipitates, and irradiation at high temperature

induced a lower density of coarse γ″ precipitates. Aging results suggest a relatively small size to establish a stable precipitate, with each precipitate only accumulating solute atoms within a small capture volume. Irradiation results suggest that solute atoms are more mobile under irradiation and can diffuse across larger distances to stable precipitates, facilitating the growth of a small density. Therefore, the hypothesis was found to be true in part, but more information will be needed at another dose or time point to determine the kinetics of the precipitation process. Several publications are in progress to describe the evolution of the microstructure, the mechanical performance with and without irradiation, and the overall processing–structure–processing relationships that can guide the use of high-strength precipitate heat treatments for Ni superalloys in nuclear environments.

The second hypothesis was to be evaluated through the tensile properties and microstructure characterization. However, although precipitate dissolution was observed after irradiation at the lower irradiation temperature, the mechanical performance was still adequate (>800 MPa yield strength) with a stronger dependence on temperature than on irradiation damage. Very few small cavities, which are indicative of He generation, were observed after one cycle in the High Flux Isotope Reactor (HFIR) and were primarily found at the lower irradiation temperature. Therefore, He embrittlement was not able to be characterized, and the hypothesis could not be evaluated as described. Specimens were irradiated in HFIR for five cycles to induce more damage and generate more He gas via transmutation. The characterization of these higher-fluence specimens may be able to refine or refute the second hypothesis.

The third hypothesis was evaluated through the comparison of precipitate size and densities measured either by human experts or an ML model. The ML model demonstrated measurements of the number of precipitates identified and their size distribution within error of the human analysis, as well as with a reduction in analysis time from several weeks to several hours. This reduction in time allowed the ML model to analyze videos of microstructure evolution during in situ ion irradiation at high temperatures in days—this same process would take a human several months of consistent analysis. Therefore, this hypothesis was proven.

10559: Mitigation of Interfacial Bottlenecks for High Energy Density Solid-State Batteries

M. Dixit

Project Description

Electrification of the transportation sector relies on radical reimagining of energy storage technologies to provide affordable, high–energy density systems that are durable and safe. Next-generation energy storage systems must leverage high–energy density electrodes and multivalent cation chemistries to achieve the required performance metrics (longer vehicle range, longer life, lower production costs, and increased safety). Solid electrolytes (SEs) are promising materials for achieving these metrics by enabling metallic anodes, but SE cells suffer from poor coulombic efficiencies and cycle life. This project aimed to study and mitigate the performance-limiting degradation mechanisms of next-generation energy storage materials used in SE cells. This work quantified electrochemomechanical dynamics via a highly versatile experimental approach to generate a comprehensive, time-resolved understanding of interfacial degradation mechanisms. Directed electrochemistry and advanced characterization techniques provided insight into the interplay of kinetics and mass transport, chemomechanics of the interface, and interfacial material microstructure. Mitigation strategies to ameliorate preempted degradation mechanisms were evaluated. This work led to a better understanding of interfacial physics that can be leveraged to design higher-performance solid-state cells and advanced performance-predicting computational models, and thus it accelerated the realization of high–energy density solid-state battery (SSB) systems.

Mission Relevance

ORNL mission directives highlight its role in supporting DOE missions of clean energy and scientific discovery. Energy storage systems are a key technology for establishing high-penetration clean energy portfolios for many applications, including grid storage systems, batteries for electric vehicles (EVs), and power supplies for manufacturing. Therefore, discovery and development of novel energy storage systems that enable high–energy density storage with safety, reliability, and economy are vital. This research project aimed to address some of the key challenges currently facing SSB technology.

Results and Accomplishments

This project showcased exemplary argyrodite and thiophosphate materials and performed solventless processing of their hybrid films with an ultraviolet-assisted curing process. The results suggested that the average adhesion and roughness of the two inorganic SE materials were similar. This finding was expected because of the similar processing conditions employed for both materials. However, the polymer hybrids of these materials showed very distinct surface morphologies, chemical makeup, and interfacial mechanical properties. Owing to the underlying subsurface distributions of the inorganic phase, the LPS composite showed much higher adhesion and a smoother surface compared with the LPSCl composite. Overall, the results supported the prospect of using polymer composites as interlayers for SSBs to regularize the interfaces in terms of surface roughness and mechanical properties. Ideally, these interlayers should be thin enough to not impede the performance of SSBs because of transport limitations.[1]

A major roadblock in the successful commercial deployment of all-solid-state batteries is the availability of a reliable, adaptable, and versatile large-scale manufacturing approach for solid-state architectures. Effectively addressing this challenge would advance present-day battery technology by enabling energy-dense SSBs to meet the burgeoning demands of portable electronics, grid storage, EVs, and even electric vertical takeoff and landing applications. The team comprehensively discussed and highlighted isostatic pressing (ISP) as a potential pathway toward large-scale production of SSBs and their components. Despite the versatility and easy adaptability of this manufacturing approach for SSBs with varying form factors, battery chemistries, and configurations, continuous R&D efforts must systematically address several underlying challenges in implementing ISP to fully leverage this manufacturing approach. To this extent, the team also summarized some key perspectives, challenges, and future directions in ISP for SSB manufacturing. (1) First, employing and optimizing ISP techniques for SSB components and cells will not only be crucial for large-scale production but also will afford key technological advantages that can improve the performance of SSBs. This point was highlighted by the ability to achieve high-density materials (>99%), the ability to control crystallographic orientations, and the ability to achieve conformal and resilient interfaces. (2) The team showed the characteristic differences between key ISP processes—cold ISP, warm ISP, and hot ISP—and their application to SSB materials and cell integration. (3) The techno-economic model discussed here provides a reference point for the scale of processing costs associated with SSB components and cells to meet future EV demands. It demonstrates that component and cell productions for SSBs become viable only with economies of scale. As innovation continues in materials and interfaces within the SSB, focusing on the manufacturing aspects of these systems is crucial. ISP processing is one of the key avenues toward achieving this goal.[2]

[1] M. Dixit et al. “Differences in the Interfacial Mechanical Properties of Thiophosphate and Argyrodite Solid Electrolytes and Their Composites.” *ACS Appl. Mater. Interfaces* 14, 39, 2022, 44,292–302.
[2] M. Dixit et al. “Tailoring of the Anti-Perovskite Solid Electrolytes at the Grain-Scale.” *ACS Energy Lett.* 7, 11, 2022, 3,936–46.

WIGNER FELLOWSHIP

10015: Direct Photons as Probes for New Forms of Quantum Chromodynamics Matter

F. Bock

Project Description

Direct photons are one of the most versatile probes in high-energy and relativistic heavy-ion physics. They can be used to measure the temperature of the quark–gluon plasma (QGP), serve as a calibration probe for energy loss measurements in the QGP, and give access to the parton distribution functions of the proton as well as ions. Their measurement is, however, challenging because of the large background coming from decays of light mesons. Improving these measurements at the Large Hadron Collider (LHC) and at a future Electron Ion Collider (EIC) will require advances in the analysis techniques and in the detector technologies used to reconstruct photons and, in particular, those close to the beam direction. This project pursued these challenges on three main fronts. On the analysis side, this work pursued advances of the measurements carried out within the ALICE (A Large Ion Collider Experiment) at the LHC in small collision systems, putting particular emphasis on the low-p_T photon reconstruction using the existing calorimeters and tracking detector. The second area investigated the further development of a 3D-imaging Forward Calorimeter (FOCal) as an upgrade of ALICE to be implemented in 2027–2030, which might enable researchers to directly observe gluon saturation within the proton and ion. To improve its performance, this project explored the installation of a charged particle veto with an integrated trigger plane to reduce the material and provide minimal particle tracking in front of the calorimeter. As a third part, this project explored whether these technologies can be adapted and improved to significantly enhance the particle identification capabilities of a future EIC detector.

Mission Relevance

In relativistic heavy-ion collisions, a strongly interacting medium with partonic degrees of freedom is formed, called an *sQGP*, offering a unique testing ground of quantum chromodynamics at high temperatures. One of the most revealing probes are photons, which are emitted at every phase of the collision and leave the expanding fireball mostly unchanged and carrying information about the medium, its evolution, and the initial conditions. Measuring the spectra and azimuthal anisotropies of direct photons at low p_T helps researchers determine the QGP's initial temperature and its time evolution. However, the exact system size and energy density required to produce a QGP still remain uncertain. A direct photon measurement in high-multiplicity p–A or p–p collisions would shed light on this question. At higher momenta, direct photons are mainly created through hard parton collisions within the proton and ion. As such, these photons give access to their substructure, about which very little is known, specifically at low fractional momenta and energy transfers. In this regime, a new state of matter could be observed, called *gluonic matter*, which can only be accessed through new instrumentation in a forward direction at existing and future colliders.

Results and Accomplishments

The focus for direct photon measurements was on reducing the systematic uncertainties—in particular, in p–p and p–Pb collisions—by tackling current instrumentation limits as well as measuring (so far, only estimated) contributions to the decay photon spectrum. In that regard, this work pushed the understanding of the ALICE electromagnetic calorimeter to a new realm of precision by accessing and publishing the Electro Magnetic Calorimeter (EMCal) performance paper,[1] reducing the related uncertainties by 30%. This project also developed a data-driven technique to correct the simulated material budget of the inner

[1] S. Acharya et al. ALICE Collaboration. "EMCal Performance Paper." *Journal of Instrumentation* 18, 8, 2023, P08007. DOI: 10.1088/1748-0221/18/08/P08007

detectors,[2] reducing the related uncertainties for the photon conversion technique to 2.5%, which increased the sensitivity to a direct photon signal at low momentum by a factor of two.

The team prepared a publication of neutral pions and eta meson as function of charged particle multiplicity in p–p collisions at $\sqrt{s}$ = 5 and 13 TeV as well as their fragmentation functions. These results lay the foundation for the first direct photon reconstruction in the same collision system with a previously unseen precision and advances the understanding of the particle production in jets. The team measured the neutral meson modification in p–Pb collisions at $\sqrt{s_{NN}}$ = 8.16 TeV,[3] which pushes the boundaries of the ALICE and allowed the team to test perturbative quantum chromodynamics and the nuclear parton distribution in a new regime. Additionally, the team measured for the first time the omega meson production, which is the third largest decay photon contribution, in p–p collisions at $\sqrt{s}$ = 5, 7[4], and 13 TeV, as well as p–Pb collisions at $\sqrt{s_{NN}}$ = 5 TeV. Beyond these reductions of uncertainties, this project also laid the foundation for future direct photon measurement using the EMCal by coordinating and implementing the new online calibration procedures necessary for the LHC Run 3 data-taking of the ALICE.

To advance the understanding of nuclear matter and the interaction within protons, this project focused on developing new detector technologies for the EIC,[5] including the design of the full calorimetry system of ePIC.[6] The principal investigator's particular focus was devoted to the forward hadronic calorimeter (HCal)—the longitudinally segmented forward HCal (LFHCal). This calorimeter is a highly integrated and highly granular Si photomultiplier (SiPM) on a tile calorimeter. The calorimeter consists of two half-disks with a radius of 2.7 m and a depth of 1.4 m. It weighs approximately 188 t. It will consist of around 600,000 individual tiles with SiPMs, which will be grouped in approximately 67,000 readout channels. It is designed to be able to handle collisions up to every 10 ns and should be able to be operated for more than 10 years total.

This work also pursued the initial idea of adding a timing layer in front of the FOCal at ALICE for ePIC. These timing layers, if installed close to the calorimeter, would allow for improved particle flow during the reconstruction as well as improved particle identification information for the charged particles hitting the calorimeters. The latter would improve the corresponding resolution because it eliminates some ambiguities during the shower reconstruction. This idea of a time-of-flight detector based on capacitively coupled low-gain avalanche diodes made out of Si with an envisioned 25–30 ps time resolution has been integrated in the current EIC detector design[7] not only in the forward but also in the full barrel acceptance.

[2] S. Acharya et al. ALICE Collaboration. "Data-Driven Precision Determination of the Material Budget in ALICE." *Journal of Instrumentation* 18, 11, 2023, P11032. DOI: 10.1088/1748-0221/18/11/P11032
[3] S. Acharya et al. ALICE Collaboration. "Nuclear Modification Factor of Light Neutral-Meson Spectra Up To High Transverse Momentum in p–Pb Collisions at $\sqrt{s_{NN}}$ = 8.16 TeV." *Physics Letters B* 827, 2022, 136943. DOI: 10.1016/j.physletb.2022.136943
[4] S. Acharya et al. ALICE Collaboration. "Production of ω Mesons in pp Collisions at $\sqrt{s}$ = 7 TeV." *The European Physical Journal C* 80, 12, 2020, 1130. DOI: 10.1140/epjc/s10052-020-08651-y
[5] J. K. Adkins et al. ECCE Consortium. "Design of the ECCE Detector for the Electron Ion Collider." Preprint, arXiv:2209.02580 [physics.ins-det], submitted to *Nuclear Instruments and Methods in Physics Research*.
[6] F. Bock et al. ECCE Consortium. "Design and Simulated Performance of Calorimetry Systems for the ECCE Detector at the Electron Ion Collider." *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 1055, 2023, 168464. DOI: 10.1016/j.nima.2023.168464
[7] J. K. Adkins et al. ECCE Consortium. "Design of the ECCE Detector for the Electron Ion Collider." Preprint, arXiv:2209.02580 [physics.ins-det], submitted to *Nuclear Instruments and Methods in Physics Research*.

10519: Closo-Borane and Carborane Functionalized Metal–Organic Framework Electrolytes for Solid-State Lithium Metal Batteries

A. Ullman

Project Description

Solid-state batteries promise higher energy densities in an inherently safe format. The key component of a solid-state battery is the solid-state electrolyte, which separates the cathode from the Li metal anode, and should enable high room-temperature Li-ion conductivity while efficiently mediating Li transport at both electrode interfaces. Polymer electrolytes are a promising class of solid-state electrolyte but generally suffer from low conductivity. Recently, it has been proposed that polymer electrolyte conductivity can be increased by (1) decreasing the electrostatic interaction between the Li cation and the counter anion and (2) introducing ion transport pathways that concentrate mobile ions in well-defined channels. This project tested this hypothesis by developing a new class of metal–organic framework (MOF) electrolytes. Weakly coordinating closo-borane and closo-monocarborane anions were incorporated into the pores of stable MOFs to create ion channels for Li transport. Experiments were designed to assess the effect on the ionic conductivity of the location, charge, flexibility, and coordination ability of the boranes. The conductivity was optimized through the judicious choice of pore-infiltrating guest molecules. MOF nanocrystals with single-ion conductivities $>10^{-3}$ S/cm were tested for their ability to mediate uniform Li metal stripping and plating.

Mission Relevance

The project addresses the DOE and ORNL missions of scientific discoveries and technical breakthroughs for applications in energy security because it pushes forward the understanding of novel materials for use in advanced Li batteries. Solid-state batteries and solid electrolytes have been the focus of several initiatives at DOE in recent years, including programs from ARPA-E and EERE's VTO. Furthermore, the materials developed during this project have potential applications in other projects that address DOE's mission, such as rare earth element metals purification and recycling and beyond-Li energy storage systems.

Results and Accomplishments

This project worked on understanding densification pressure effects during MOF electrolyte membrane processes. The team used two processing methods—a dry process using polytetrafluoroethylene as the binder and a wet process using polyvinylidene fluoride as the binder—to fabricate MOF membranes using a model MOF material, MOF-808. The effect of fabrication pressure on the crystallinity, surface area, and microporosity as well as transport properties of the resulting pellets and membranes were thoroughly examined. The porosity and liquid electrolyte uptake are a strong function of fabrication pressure. As the pressure increased, the porosity decreased. In contrast, the conductivity of the membranes is not a strong function of fabrication pressure. It is expected that these findings will be generalizable to the processing of other MOF materials in which porosity is a crucial factor for battery performance.

Further elucidation of the connection between the macroscopic application of pressure and microscopic intra- or interparticle ion transport has been the focus of ongoing work that uses MOF-808 particles with covalently tethered anions. The synthesis of the functionalized MOF-808 variants has involved the incorporation of 4-sulfobenzoic acid into the pores of the MOF. Although ^{1}H nuclear magnetic resonance analysis of the digested MOF has shown high levels of substitution of the anion, recent ^{9}Li nuclear magnetic resonance experiments have demonstrated that the sulfonate anion is not paired with a Li cation despite careful washing with LiCl. New ion exchange techniques are now being pursued in which pH is controlled during washing. This work has also discovered that structural linkers with functional groups can be postsynthetically incorporated onto the outer surface of the MOF particles. The team is currently using these functionalized MOFs for new membranes that feature covalent attachments between the MOF

particle and the polymer binder. The team expects that membranes made in this way will have improved interfacial stability and therefore cycle longer in a Li metal battery.

10554: Quantum Computing Applications for High-Energy Physics

A. Delgado

Project Description

Quantum chromodynamics (QCD) is the theory of strong interactions, which are the fundamental forces in nature that binds quarks and gluons into hadrons, such as protons and neutrons. Although the Higgs particle gives mass to the quarks, only a tiny fraction of that mass accounts for the mass of protons, neutrons, and their partner particles. QCD provides mass to these composite objects. The study of QCD properties is one of the central scientific challenges in both nuclear and high-energy physics. Currently, QCD has not been solved exactly, and some calculations and numerical approximations that shed light on nuclear masses and their interactions involve complex computational methods and large-scale simulations. These simulations often involve the use of state-of-the-art computational resources. A potential alternative is harnessing the quantum mechanical nature of matter to enhance or accelerate scientific goals and discoveries through a more suitable computing technology—quantum computing. This project aimed to expand the current understanding of QCD interactions using quantum computing to perform accurate calculations of interest in nuclear physics and high-energy physics. Specifically, this work attempted to improve current jet properties predictions, which are essential tools for discovering new physics and understanding and testing perturbative and nonperturbative QCD at collider experiments. This goal was achieved by using quantum computing to study the quark–gluon cascade's evolution after QCD confinement is broken during high-energy collisions.

Mission Relevance

This work supported the effort of identifying quantum information science solutions to high-energy physics and nuclear physics computing challenges identified by DOE.

Results and Accomplishments

This proposal addresses how to interface data from realistic physical models and quantum computing experiments. This interfacing is usually a bottleneck in the overall workflow because the potential speedup obtained using quantum computing to perform a calculation on classical data is often shadowed by the computational burden of transforming bits from the classical domain to quantum states. This often inefficient process becomes a shortcoming for large sets of classical data, and this must be taken into account when reporting any potential quantum advantage.

The technical approach relied on quantum algorithms based on quantum annealing processors, such as the D-Wave processors, that are currently available through the Oak Ridge Leadership Computing Facility Quantum Users Program. In quantum annealing, the solution to an optimization problem is encoded in a target Hamiltonian's ground state. This target Hamiltonian takes the form of an Ising model in which spins correspond to qubits, and the user programs the weights and couplings between qubits. A transformation from spin to binary variables frames the optimization problem as a quadratic unconstrained binary optimization (QUBO), which is solved via quantum annealing. This study implemented and validated the jet-clustering algorithms. These algorithms provide a quantum-alternative to the classical jet-finding algorithms. Identifying jets formed in high-energy collisions requires (classically) solving optimization problems over potentially large numbers of final-state particles. Focusing on the relatively simple case of electron–positron collisions, this work considered a well-known event shape called *thrust*. In high-energy physics, thrust is a jet substructure variable that characterizes the event shape. An event with spherically distributed particles would have a thrust value equal to 0.5, and a two-jet event would have a thrust value closer to unity. Such a variable's optimum corresponds to the most jet-like separating plane among a set of particles, thereby defining two hemisphere jets. In this

sense, thrust can be calculated for a candidate partition as a QUBO problem. This formulation can be seen as a partition problem in which particles on either side of the partition constitute a jet.

The dataset of electron–positron events was be generated using Pythia 8 and benchmarked against the performance of the classical implementation of both the e + e− anti-kT and SingleCone algorithms. These two classical algorithms correspond to the cone-finding and hierarchical clustering approaches, respectively. Usually, the cone-finding approach type is preferred to study the constituent particles' phenomenological properties, but the hierarchical clustering approach is preferred when CPU performance is considered. This project compared the physics and CPU performance of the classical formulation with the quantum counterpart. The characterization of the general phenomenological properties of jets identified using the QUBO formulation for thrust was completed. This study involved direct comparison with Pythia's thrust-finding algorithm, which yields an axis that partitions the jet phase space into two jets. The QUBO jet finder, on the other hand, has an effective jet radius that depends on the mass of the jet. This study concluded that the QUBO jet finder was able to perform the clustering task as efficiently as its classical counterpart when solved through the classical heuristic algorithm, simulated annealing, but the performance of the D-Wave solver was inferior when compared with its classical counterpart. The results were published in a peer-reviewed journal article.[8]

Analysis and simulation of hadronic processes will be a crucial task in current and future high-energy physics experiments. The process of simulating the detector response is usually computationally expensive, especially for QCD processes. This project aimed to develop a quantum machine learning (QML) method to speed up the current algorithms for particle shower simulation in high-energy physics experiments. A *particle shower* is a cascade of secondary particles produced when an identified primary particle interacts with a detector. In a particle shower, the initial particle decays or produces multiple new particles with lesser energy, which eventually interact with the detector and produce even more new particles. This study used hierarchical machine learning methods and quantum-classical neural networks to tune the training complexity for generative models of particle showers. These QML methods for the enhanced simulation of particle showering and detector response will enable high-fidelity, fast simulations of the kinematics of a particle in a typical proton–proton collision by offering a solution to the difficult challenge of simulating a longitudinally segmented calorimeter because of the sparsity of cells, the nonuniform granularity among the detector layers, and their subsequent structure. This approach was to train a quantum circuit Born machine as a quantum-assisted generative model specifically for high-energy physics applications. The end goal was to employ QML methods to generate particle kinematic distributions from a model trained using classical Monte Carlo simulations. The quantum circuit Born machine was expected to correctly model and capture the nonlinear functional representations of the experimental observables' simulated data distribution.

In summary, the project execution plan included two main objectives dealing with the development, application, and benchmarking of quantum computing algorithms applied to typical high-energy physics data analysis tasks and understanding which areas of experimental high-energy physics will be benefited the most from quantum algorithms executed on near-term devices. The first objective dealt with developing quantum-assisted algorithms for jet clustering and track reconstruction in proton–proton collisions. The study allowed for understanding the limitations of current devices to process the amount of data collected by a typical particle physics experiment. The second thrust dealt with the generation of quantum generative models to harness the capabilities of quantum processors to sample from high-dimensional feature space and augment the statistics of datasets generated with classical Monte Carlo techniques. A generative model suitable for the generation of kinematic distributions of particles in a

[8] A. Delgado and J. Thaler. "Quantum Annealing for Jet Clustering with Thrust." *Physical Review D* 106, 2022, 094016. DOI: 10.1103/PhysRevD.106.094016

typical proton–proton collision at a hadron collider was developed and benchmarked on IBM hardware. Project results are available in a publication.[9]

[9] A. Delgado and K. E. Hamilton. "Unsupervised Quantum Circuit Learning in High Energy Physics." *Physical Review D* 106, 2022, 096006. DOI: 10.1103/PhysRevD.106.096006

SUMMARIES OF PROJECTS SUPPORTED BY THE EARLY CAREER COMPETITION

NEUTRON SCATTERING DIVISION

11348: Unlocking In-Situ Quantum Magnetism Engineering with Neutrons

K. Taddei

Project Description

This project aimed to establish techniques for quantum magnetism engineering with the goal of accessing technologically relevant quantum phases of matter using selectively tuned pulsed lasers to disrupt magnetically ordered materials and quench targeted magnetic interactions. This method leaves the materials to order under theoretically ideal interaction Hamiltonians and thus stabilize quantum magnetic phases such as the Kitaev quantum spin liquid or the Haldane chain. These intensely studied phases theoretically give rise to exotic magnetic states with long-range quantum entanglement and novel quasi particle excitations highly sought for quantum information applications. However, the desired physics are often obscured by weakly perturbing interactions in the material, impeding their study and implementation. This project envisioned a pulsed laser being used to weaken these perturbing interactions using the new concept of a *time window mismatch* quench, leading to more purely quantum magnetic states. This method would allow for the experimentally controlled tuning of magnetic interactions in candidate materials known to be *proximate* to quantum magnetic states, driving them into the theoretically ideal configurations. This project was intended to be done in situ on neutron scattering beamlines using the magnetic information accessed in neutron scattering to study the stabilized quantum magnetic phases and understand the quenching mechanism. The access to quantum magnetic states provided by this project would elucidate the functionality of these phases and give a deeper understanding of the underlying physics.

Mission Relevance

Quantum materials have a singular potential to revolutionize the technological landscape from quantum computing and sensing to radically new forms of energy materials. New robust material platforms for the qubit are necessary to achieving scalable quantum computers, which are important for national security. Quantum devices based on the exotic physics derived from quantum materials more generally may enable new forms of highly efficient photovoltaics, revolutionary new catalysts, and instantly charging batteries. However, this promise hinges on the ability to stabilize, study, and optimize the relevant quantum and topological phases. With this ability to contribute to both national security and clean energy, this project aligns with DOE’s mission. It will work to build the experimental capacity to tune materials into these phases, thus helping to accelerate the ability to unlock their potential.

Results and Accomplishments

This project ended early.

INDEX OF PROJECT NUMBERS