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2024 Annual Report Laboratory Directed Research & Development

March 2025

Changing the World's Energy Future

Tony Huff, Stephannie A Lambert

Idaho National Laboratory

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Idaho National Laboratory
Idaho Falls, Idaho 83415

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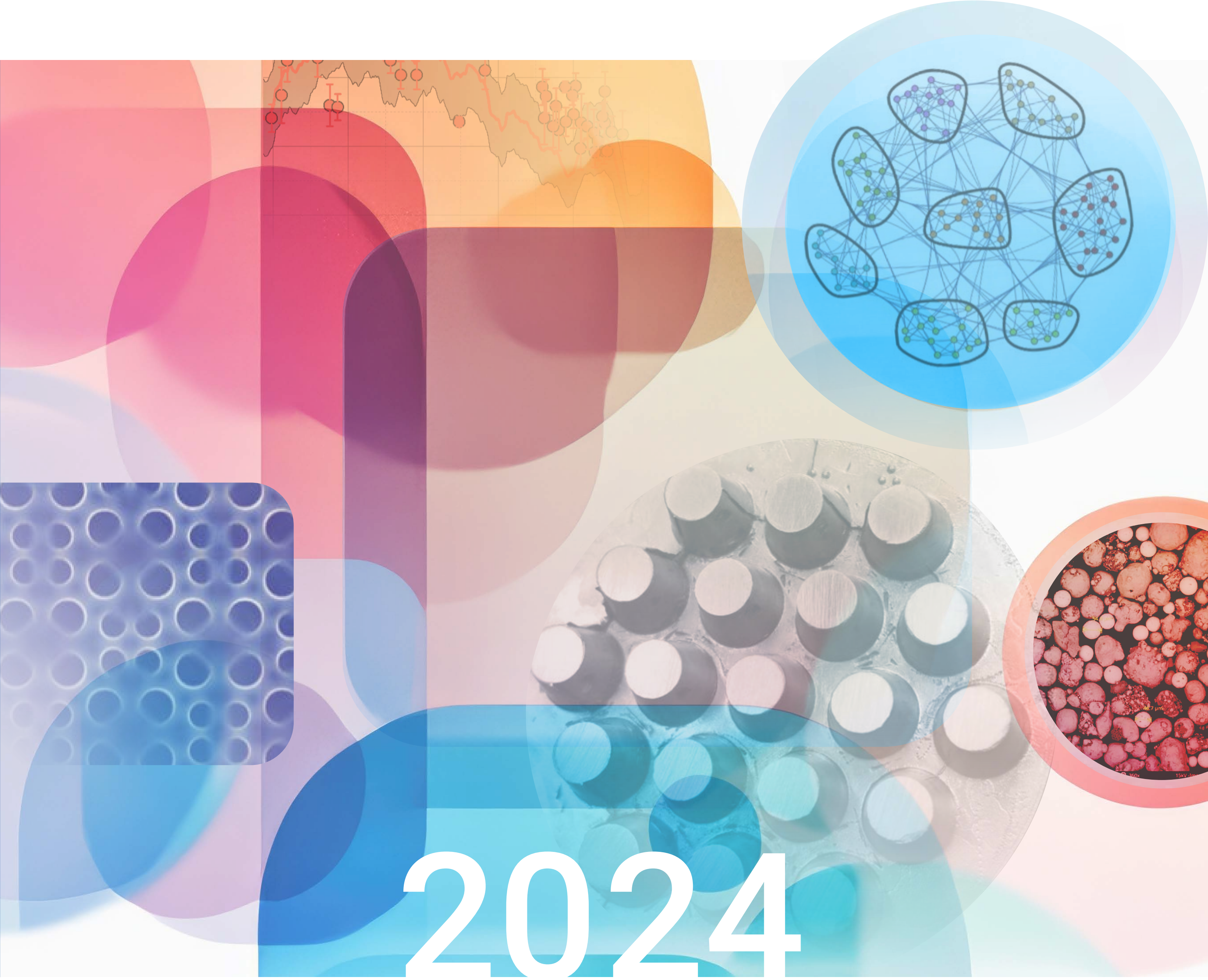

2024

ANNUAL REPORT

Laboratory Directed Research & Development

Idaho National Laboratory

Celebrating 75 years of Innovation

FROM INL'S CHIEF RESEARCH OFFICER

DR. TODD COMBS
Deputy Laboratory Director for Science and Technology and Chief Research Officer, Idaho National Laboratory

I am pleased to present the fiscal year 2024 (FY-24) Laboratory Directed Research and Development (LDRD) Annual Report for Idaho National Laboratory (INL). This report highlights some of the innovative and impactful research projects that have been pursued over the past year through the LDRD program, which remains a cornerstone of our commitment to scientific excellence and technological innovation.

The LDRD program is instrumental in driving the frontier of research, enabling us to explore high-risk, high-reward scientific endeavors that align with INL's strategic objectives and national priorities. This year, our researchers made significant strides in areas such as nuclear energy, cybersecurity, integrated energy systems, and advanced materials. Their work advances our understanding of critical scientific questions and paves the way for groundbreaking technologies that will shape the future of energy and security.

In FY-24, we have seen remarkable progress and achievements across our LDRD portfolio. These projects have fostered innovation within INL and have facilitated collaborations with academic institutions, industry partners, and other national laboratories. Such partnerships are vital for leveraging diverse expertise and resources, ultimately enhancing the impact of our research.

This research was conducted by a diverse group of researchers, technicians, and support staff whose dedication and hard work have made these accomplishments possible. Their commitment to excellence and innovation is what drives our success and propels us forward.

As we look ahead, the LDRD program will continue to be a catalyst for scientific discovery and technological innovation at INL. We remain committed to pursuing bold and visionary research initiatives that support our mission to discover, demonstrate, and secure innovative nuclear energy solutions, other clean energy options, and critical infrastructure.

Thank you for your continued interest, support, and engagement with INL's research mission. I invite you to explore the projects in this report and review the achievements of our talented research community.

With warmest regards,
Todd Combs

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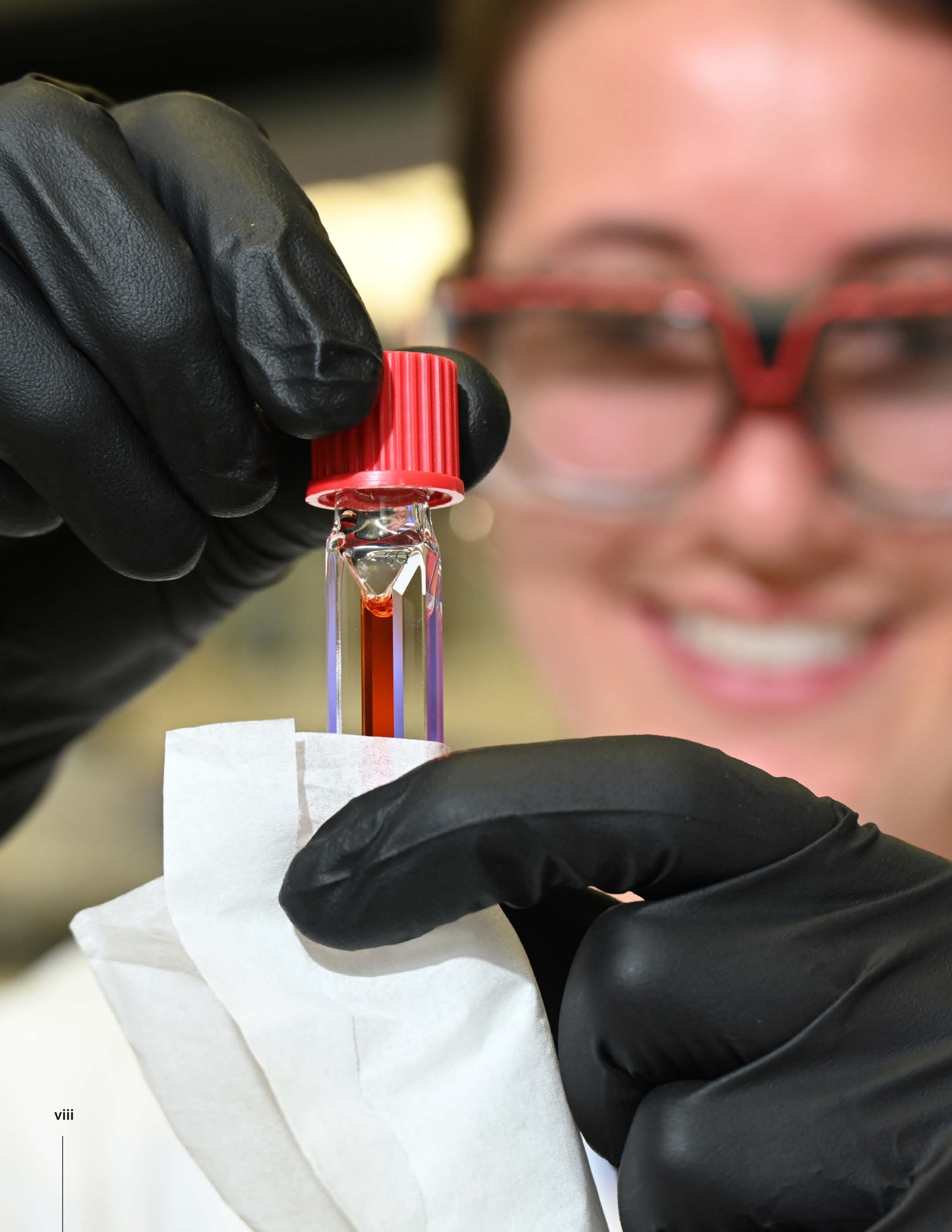
Nuclear Reactor Sustainment and Expanded Deployment

Integrated Fuel Cycle Solutions

Integrated Energy Systems

Advanced Materials and Manufacturing for Extreme Environments



LDRD OVERVIEW

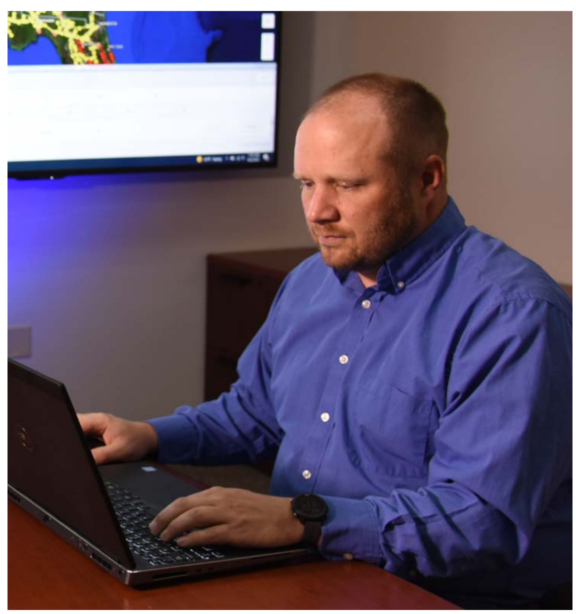
In 1991, the United States (US) Congress approved a Department of Energy (DOE) recommendation to establish the LDRD program as a means for the national laboratories to devote a small portion (maximum of 6%) of their research effort to creative and innovative work to maintain their scientific and technical vitality in disciplines relevant to energy and national security missions. The LDRD program serves as a proving ground for advanced research and development concepts that may not be otherwise pursued through direct programmatic funding sources.

DOE LDRD OBJECTIVES

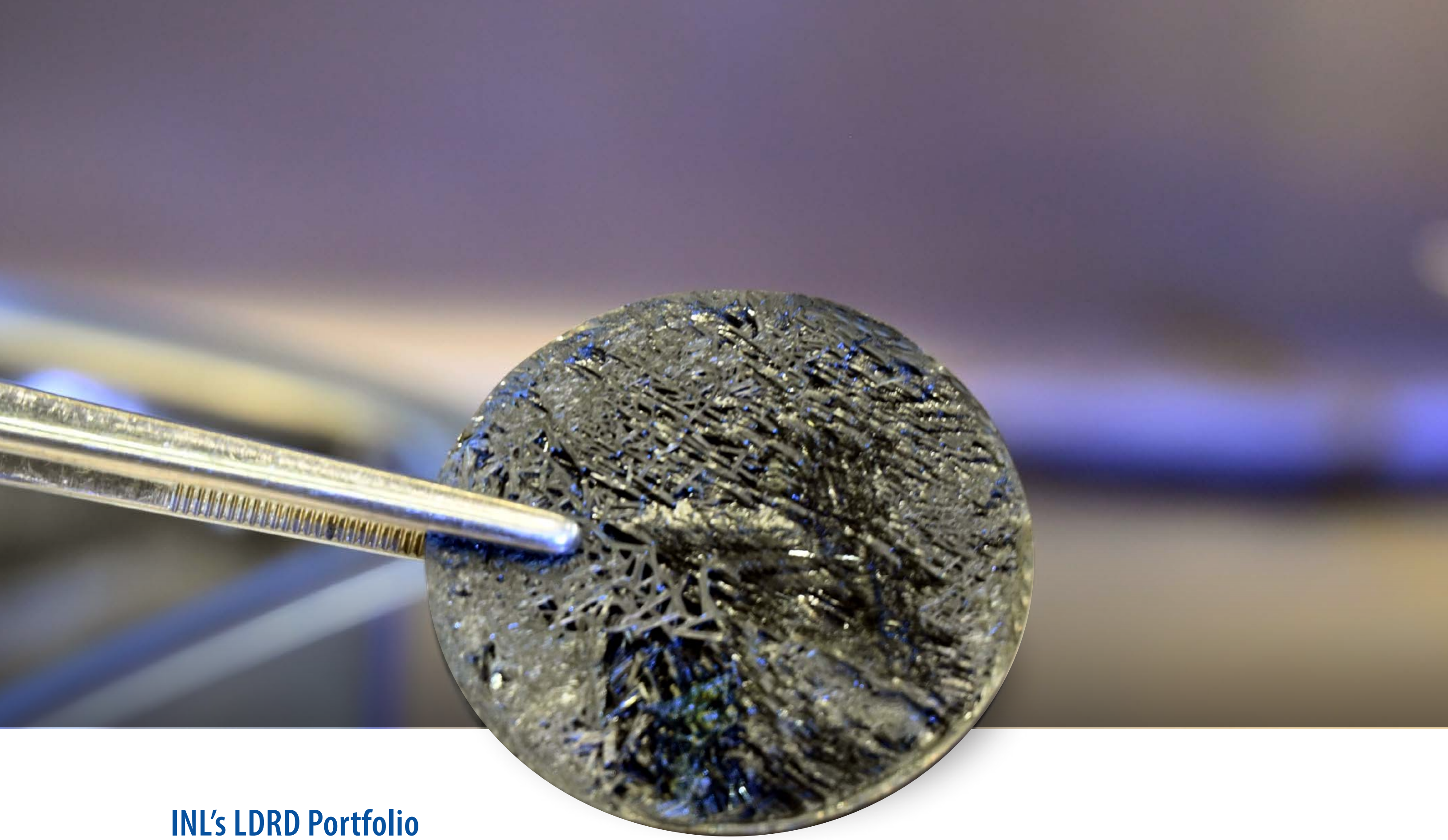
SCIENTIFIC AND TECHNICAL VITALITY
Advance the frontiers of science, technology, and engineering.

WORKFORCE DEVELOPMENT
Attract, retain, and develop tomorrow's scientific and technical workforce.

MISSION AGILITY
Enable agile responses to national security, energy, and environmental challenges.



RYAN HRUSKA



INL's LDRD Portfolio

INL's diverse LDRD portfolio explores a range of scientific and engineering concepts through technically sound, innovative, and novel research projects. The LDRD program stimulates exploration in basic and applied science and engineering. The LDRD portfolio comprises five investment components that are continually aligned with INL's vision, mission, and science and technology initiatives.

- The strategic research and development fund supports research that advances INL's science and technology initiatives.
- The seed fund supports high impact, innovative research that is aligned with INL's mission, even if not explicitly aligned with a science and technology initiative.
- The distinguished postdoc fund supports early career researchers in INL's three distinguished postdoctoral fellowships, providing them leadership opportunities while they conduct leading edge research that supports INL's mission.
- The strategic hire fund grows INL science and technology capability by providing new hires opportunities to conduct research aligned with our mission.
- New in FY-24, the early career researcher fund promotes the careers and future opportunities available to INL's early career research staff working in areas of interest to DOE Office of Science. These projects are funded for one fiscal year. Each principal investigator has a mentor who helps them navigate an Office of Science early career award submittal following the LDRD project. In this inaugural year, five early career researcher LDRD projects were funded.

» Dr. Alexander Lindsay's project "Hybridized discontinuous Galerkin methods for computational fluid dynamics" is on page 33.

» Dr. Meng Li's project "Revealing small ion intercalation dynamics through data mining of trajectories" is on page 104.

» Dr. Abderrahman Atifi's project "Rationalizing the role of f-orbitals coordination in the rare earth metals electrodeposition in organic electrolytes" is on page 106.

» Dr. Brelon May's project "Breaking symmetry to induce unconventional superconductivity" is on page 126.

» Dr. Amey Khanolkar's project "Ultrafast charge carrier-phonon interaction and relaxation dynamics in transition metal nitrides under pressure" is on page 124.

Top photo: INL researchers synthesizing Zero Power Physics Reactor highly enriched uranium into salt (sodium chloride-uranium trichloride eutectic). This is fuel salt for the LDRD-funded Molten Salt Research Temperature Controlled Irradiation (MRTI). This fuel salt will be irradiated in NRAD and then post-irradiation examination will happen in the Molten Salt Thermophysical Examination Capability.

$53 MILLION TOTAL PROJECT BUDGET UP FROM $47M MILLION IN FY-23.

The INL LDRD program budget increased from $47M in FY-23 to $53M in FY-24. Of the 149 active projects in FY-24, 62 concluded and are highlighted in this report. To increase the value and impact, the program expects that researchers should have at least one peer reviewed publication for each $200k of LDRD project funding. This is part of INL's overall strategy to continue strengthening its basic and applied research excellence culture. Intellectual property generation is also important to INL's research excellence, including invention disclosures, software disclosures, patent applications, copyrights, and licenses.

Core Capabilities

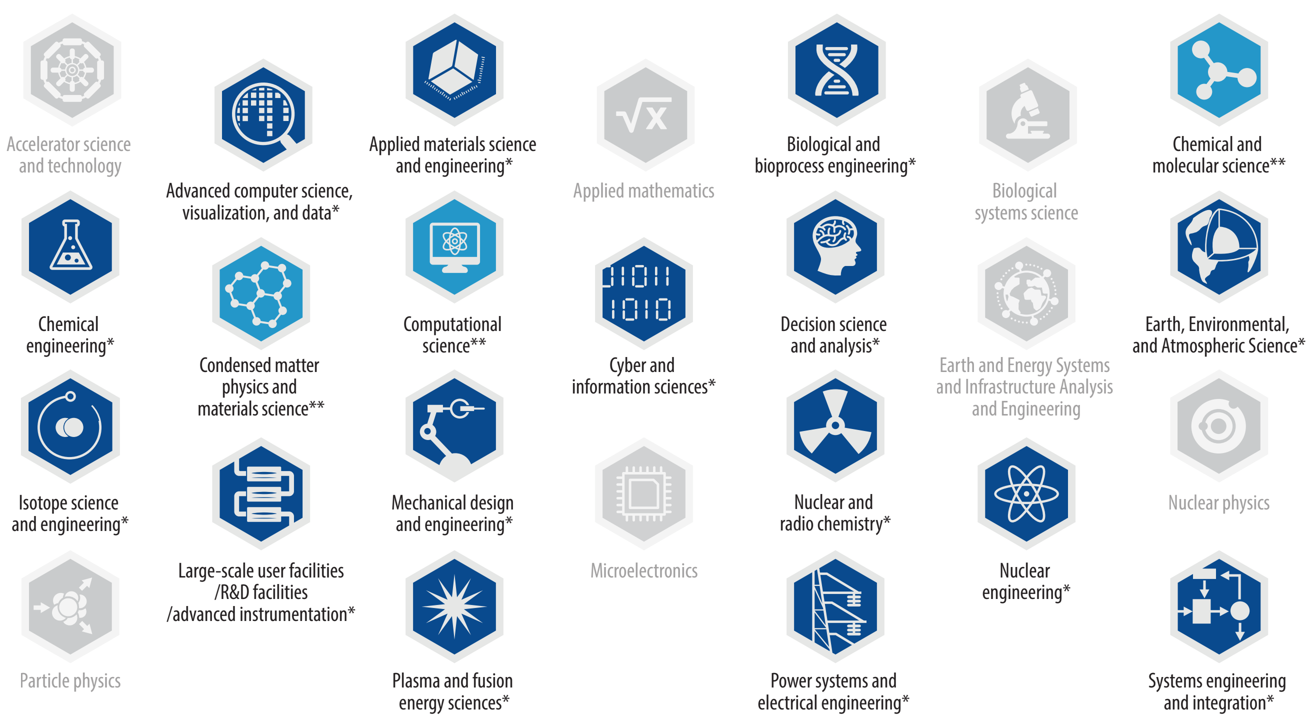
INL creates a discovery and solution driven research, development, and demonstration (RD&D) environment through unique experimental systems, infrastructure, and fundamental engineering and science capabilities. Of the 25 core capabilities foundational to DOE's mission and shared across the national laboratories, DOE acknowledges that INL has 15 core capabilities and three emerging core capabilities. These core capabilities represent a comprehensive science and engineering skill set that extends across a continuum, connecting basic and applied research to develop, test, demonstrate, and validate technologies at scale, speeding deployment and reducing risks.

These core capabilities are sustained and enhanced through INL's LDRD projects. To demonstrate that the emerging core capabilities are intrinsic at INL, the LDRD program included the three emerging core capabilities as strategic scientific initiatives. Of the 62 projects highlighted in this report, 32 demonstrated one or more emerging core capabilities.

Throughout this report, icons will indicate the core capabilities supported by LDRD projects.

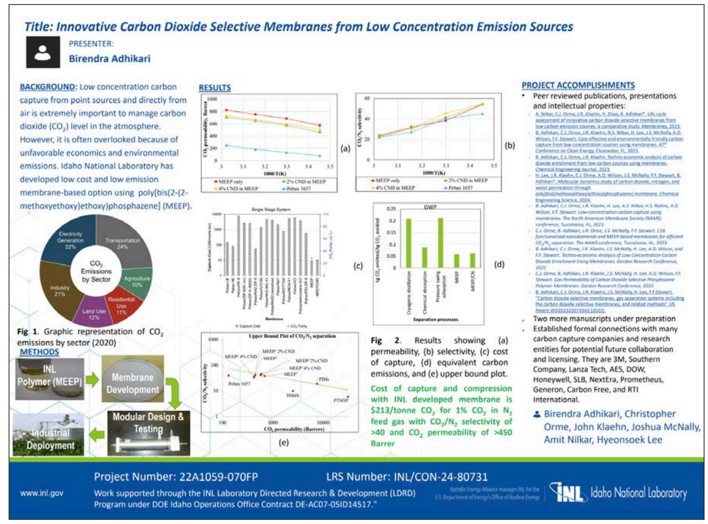
Project Selection & Oversight

INL ensures that LDRD program goals and objectives are aligned with DOE Order 413.2C, Chg. 1, and that the LDRD portfolio is managed with integrity and transparency. Project proposals and progress reports are subject to multiple levels of rigorous review by subject matter experts and senior leaders. The deputy laboratory director for science and technology reviews projects recommended for approval with the associate laboratory directors and makes final funding decisions on the LDRD portfolio. Finally, DOE Idaho Operations Office concurrence is required on each proposal and project continuing to the next fiscal year prior to project funding.



Core Capabilities *INL (dark blue) ** Emerging (light blue)

Dr. Mohammad Abdo's project on "Promoting optimal sparse sensing and sparse learning for nuclear digital twins" won first place for best poster.



Dr. Birendra Adhikari won second place for "Innovative carbon dioxide selective membranes from low concentration emission sources."

Showcasing Success

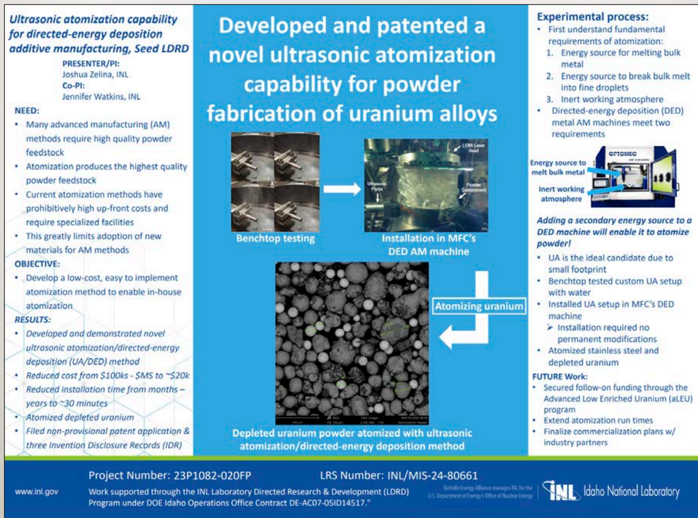
On September 23, 2024, INL hosted a poster session and symposium showcasing the LDRD projects ending in FY-24. Attendees included INL researchers, industry and academic partners, external collaborators, and members of the public. Images of the posters are available on INL.gov.

62 LDRD PROJECTS WERE FEATURED AT THE POSTER SESSION AND ARE HIGHLIGHTED IN THIS REPORT.

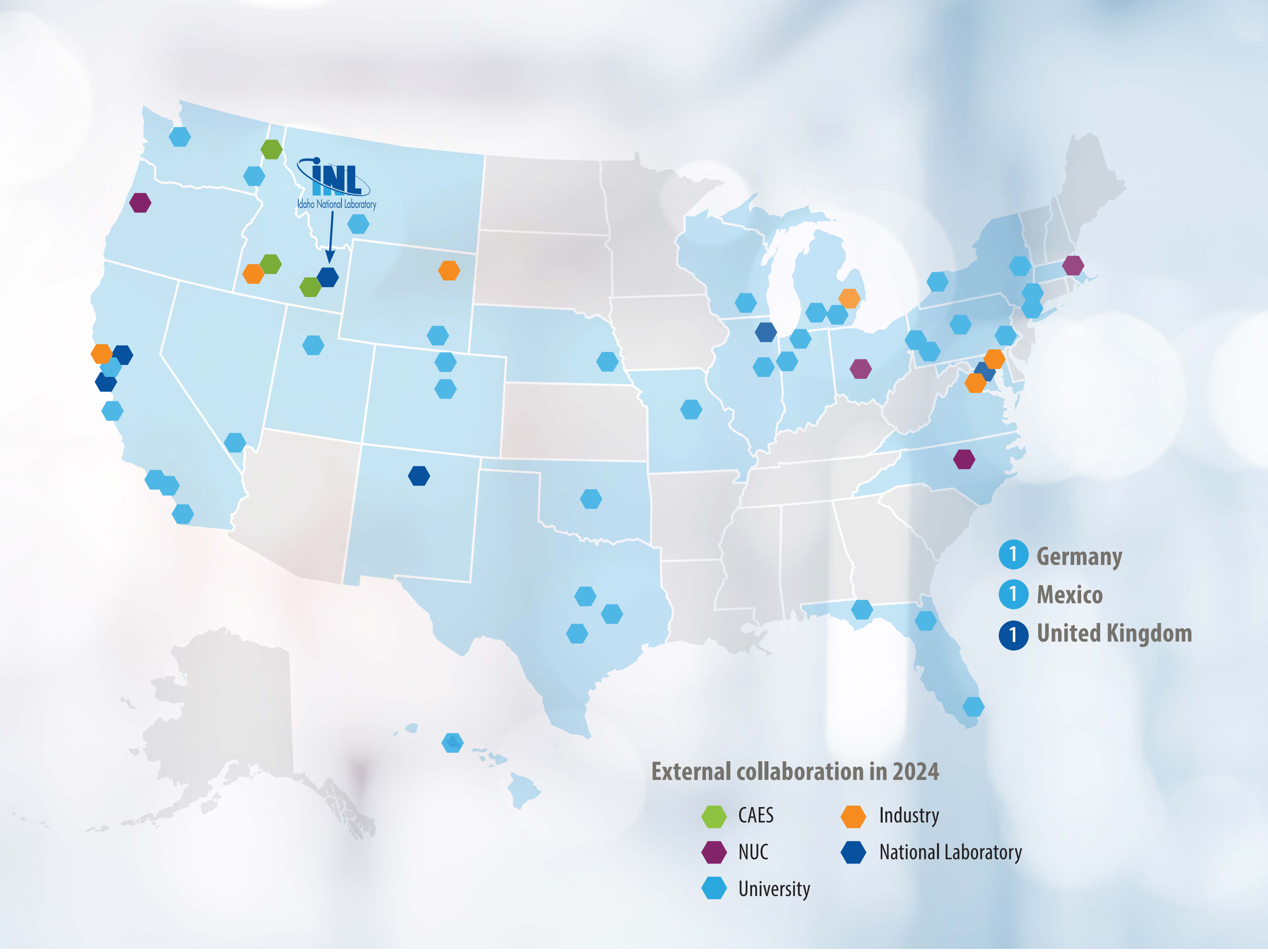
The poster session featured all 62 projects highlighted in this report. In a "best poster" competition, posters were evaluated on their creativity addressing cutting edge science, technology, or engineering, the significance and impact of the project outcomes, the presentation clarity, and the potential for attracting future RD&D funding opportunities. Dr. Mohammad Abdo's project on "Promoting optimal sparse sensing and sparse learning for nuclear digital twins" summarized on page 20 won first place for best poster. Dr. Birendra Adhikari won second place for "Innovative carbon dioxide selective membranes from low concentration emission sources." See the project summary on page 42. Mr. Joshua Zelina won third place for "Ultrasonic atomization capability for directed-energy deposition additive manufacturing," which is summarized on page 75.

The symposium featured ten oral presentations spanning all five science and technology initiatives and three emerging core capabilities chemical and molecular science, computational science, and condensed matter physics and materials science. Additionally, Jason Stolworthy, director of INL's Technology Deployment Office, presented the symposium keynote on "Maximizing the Impact of Your LDRD with Innovation."

Sample array featuring 19 unique high entropy alloy compositions produced simultaneously using the newly developed parallelized electric field assisted sintering process.



Dr. Joshua Zelina won third place for "Ultrasonic atomization capability for directed-energy deposition additive manufacturing."



COLLABORATION

INL's LDRD program encourages collaboration across organizational, institutional, and geographical boundaries to advance the frontiers of science, technology, and engineering. Of the approximately 440 researchers working on LDRD projects in FY-24, 110 of them were outside INL and represented 27 states as well as Germany, Mexico, and the United Kingdom. These external collaborators included 94 researchers from 46 universities, ten researchers from six national laboratories, and six researchers from different companies.

110 RESEARCHERS OUTSIDE OF INL COLLABORATED ON LDRD PROJECTS.

The Center for Advanced Energy Studies and the National University Consortium facilitate collaboration with particular research universities to further INL's mission. The Center for Advanced Energy Studies is a research and education consortium consisting of INL and the public research universities of Idaho: Boise State University, Idaho State University and University of Idaho. The National University Consortium includes the partner universities Massachusetts Institute of Technology, North Carolina State University, The Ohio State University, Oregon State University and University of New Mexico. Twenty-six researchers from these consortia were co-investigators on LDRD projects in FY-24, and many more students and postdocs contributed.

68 POSTDOCS FROM 50 INSTITUTIONS

121 INTERNS FROM 67 INSTITUTIONS

46 UNIVERSITY COLLABORATORS

11 NONPROVISIONAL U.S. PATENT APPLICATIONS

28 INVENTION DISCLOSURE RECORDS

8 PATENTS GRANTED

2 COPYRIGHT ASSERTED

98 PUBLICATIONS

15 SOFTWARE DISCLOSURE RECORDS

55 LICENSES ISSUED

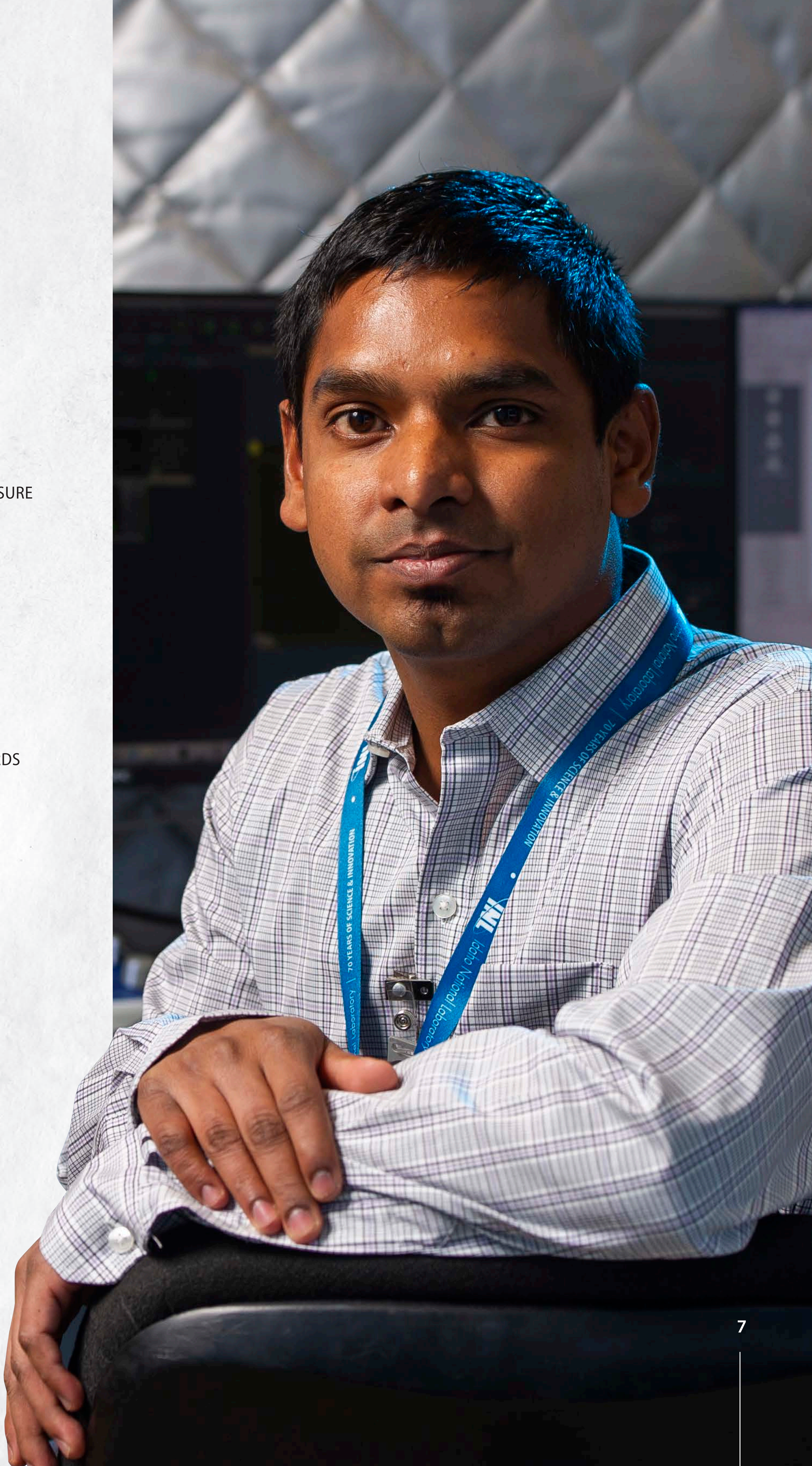
66 NEW PROJECTS

62 PROJECTS ENDING

149 ACTIVE PROJECTS

6 INDUSTRY COLLABORATORS

6 DOE OR OTHER FEDERAL LAB COLLABORATORS



KAUSTUBH BAWANE

ACRONYMS & ABBREVIATIONS

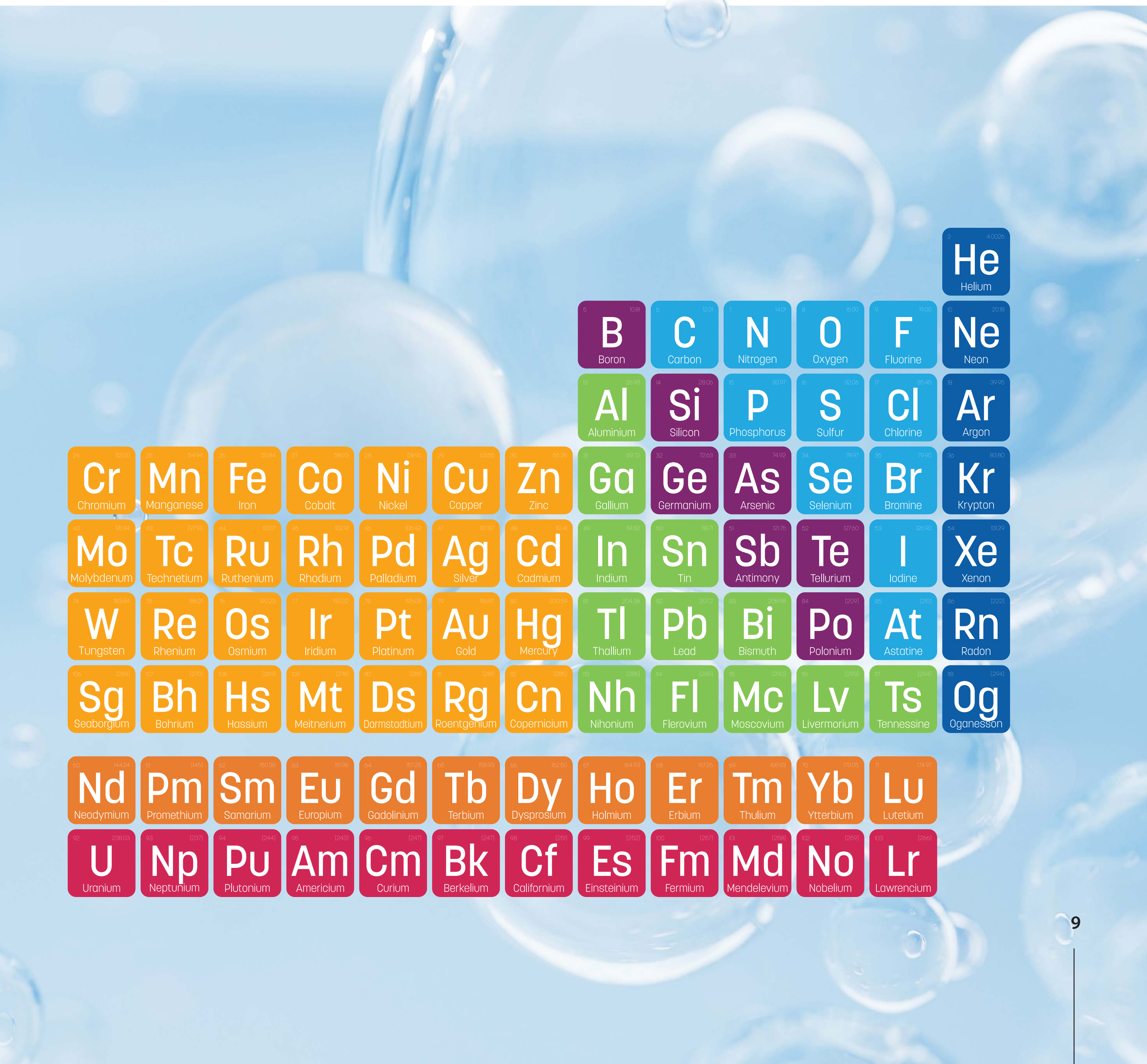
DOE Department of Energy

INL Idaho National Laboratory

LDRD Laboratory Directed Research and Development

MOOSE Multiphysics Object-Oriented Simulation Environment

RD&D Research, development, and demonstration


He Helium
B Boron
C Carbon
N Nitrogen
O Oxygen
F Fluorine
Ne Neon
Al Aluminium
Si Silicon
P Phosphorus
S Sulfur
Cl Chlorine
Ar Argon
Cr Chromium
Mn Manganese
Fe Iron
Co Cobalt
Ni Nickel
Cu Copper
Zn Zinc
Ga Gallium
Ge Germanium
As Arsenic
Se Selenium
Br Bromine
Kr Krypton
Mo Molybdenum
Tc Technetium
Ru Ruthenium
Rh Rhodium
Pd Palladium
Ag Silver
Cd Cadmium
In Indium
Sn Tin
Sb Antimony
Te Tellurium
I Iodine
Xe Xenon
W Tungsten
Re Rhenium
Os Osmium
Ir Iridium
Pt Platinum
Au Gold
Hg Mercury
Tl Thallium
Pb Lead
Bi Bismuth
Po Polonium
At Astatine
Rn Radon
Sg Seaborgium
Bh Bohrium
Hs Hassium
Mt Meitnerium
Ds Darmstadtium
Rg Roentgenium
Cn Copernicium
Nh Nihonium
Fl Flerovium
Mc Moscovium
Lv Livermorium
Ts Tennessine
Og Oganesson
Nd Neodymium
Pm Promethium
Sm Samarium
Eu Europium
Gd Gadolinium
Tb Terbium
Dy Dysprosium
Ho Holmium
Er Erbium
Tm Thulium
Yb Ytterbium
Lu Lutetium
U Uranium
Np Neptunium
Pu Plutonium
Am Americium
Cm Curium
Bk Berkelium
Cf Californium
Es Einsteinium
Fm Fermium
Md Mendelevium
No Nobelium
Lr Lawrencium

NUCLEAR REACTOR SUSTAINMENT AND EXPANDED DEPLOYMENT

As the nation's nuclear energy RD&D laboratory, our technical leadership advances US global competitiveness by sustaining the safe and efficient operation of existing reactors and pioneering advanced nuclear energy technologies for future deployment. Our innovative technologies will improve the performance of existing and future nuclear energy systems, including nuclear fuels and materials, and we will demonstrate two first-of-their-kind advanced reactors and one advanced reactor experiment over the next three years. We will expand and advance current nuclear infrastructure, experimental, and modeling and simulation capabilities. We will apply our extensive nuclear expertise to discover and develop innovative RD&D solutions in fusion blanket technologies, while helping build the workforce, infrastructure, and partnerships necessary to design and test integrated components of fusion power systems.

CORE CAPABILITIES

Applied Materials Science & Engineering

Applied Mathematics

Computational Science

Condensed Matter Physics and Materials Science

Decision Science and Analysis

Isotope Science and Engineering

Nuclear and Radio Chemistry

Nuclear Engineering

Systems Engineering and Integration

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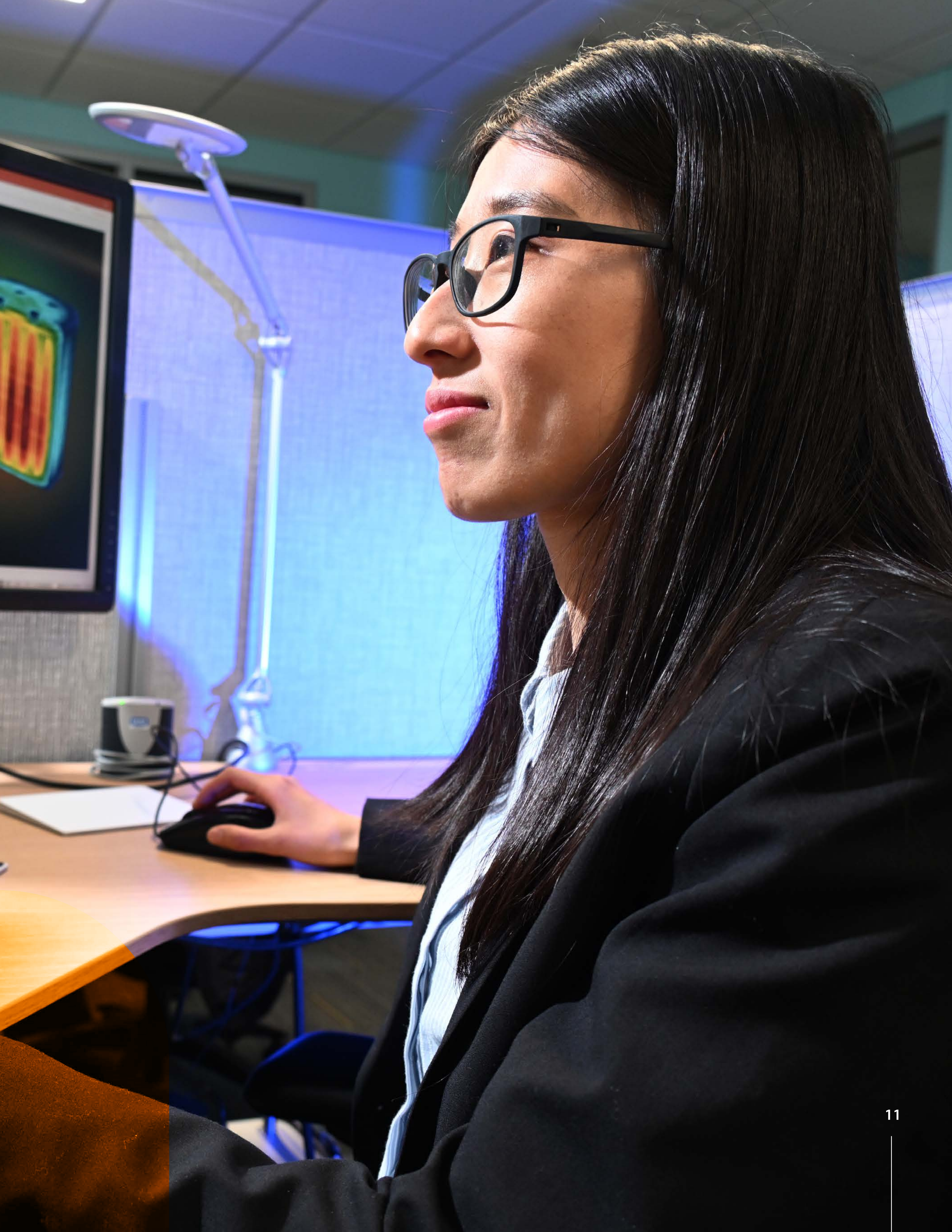


Design and implementation of the experimental setup of the three-omega method for thermal conductivity measurements of molten actinide salts

PROJECT NUMBER:
21P1056-013FP

TOTAL APPROVED AMOUNT:
$487,700 over 3 years

PRINCIPAL INVESTIGATOR:
Maria del Rocio Rodriguez Laguna

An innovative custom-built three-omega setup shows promising thermal conductivity results for solids and liquids at room temperature and is now ready for high-temperature testing.

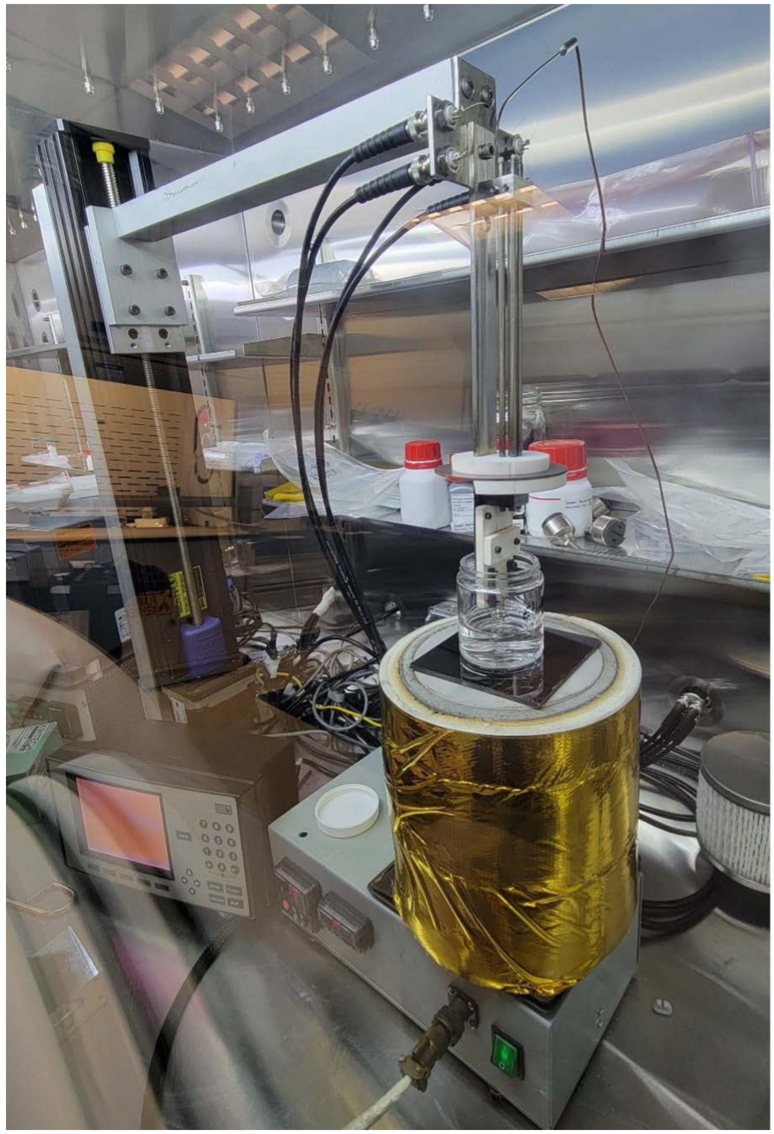
We successfully developed an innovative thermal conductivity sensor and experimental setup based on the three-omega method. The setup is designed to withstand high temperatures and corrosive molten salts while accurately measuring the thermal conductivity of liquids without convection interference. To effectively design, model, predict, license, and operate molten salt reactors, it is imperative to comprehend the behavior of both the fuel and the coolant in relation to temperature. Measuring thermal conductivity in molten salts poses various challenges, including (i) the need to operate at elevated temperatures, (ii) the corrosive nature of molten salts, and (iii) the presence of competing heat transfer mechanisms that can potentially disrupt precise measurements. To overcome these obstacles, we employed the three-omega technique, which enables rapid measurements and mitigates interference caused by convection. This characteristic plays a pivotal role in ensuring accurate measurements and underscores the superiority of this technique over alternative approaches. Additionally, we developed sensors that exhibit high-temperature resilience and can withstand corrosive environments. We successfully developed and tested the sensor, designed and validated the circuit, and achieved full automation of the setup. The thermal model has been validated for accurate measurements of solids at room temperature and up to 120°C, yielding results consistent with literature values. Additionally, the setup has demonstrated promising results for liquid measurements at room temperature and is now ready for high-temperature testing.

TALENT PIPELINE:

- Maria del Rocio Rodriguez Laguna, *Glenn T. Seaborg distinguished postdoctoral fellow at INL*, converted to staff

PRESENTATION:

- Rodriguez-Laguna, M.R., "Design and implementation of an experimental setup based on the three-omega method for thermal conductivity measurements of molten salts," The Minerals, Metals and Materials Society, March 19-23, 2023.



Three-omega setup inside a glovebox. The sensor is immersed in ethylene glycol during a room temperature measurement. The furnace is used for high-temperature measurements. Instruments are located outside the glovebox, and the system control is automated.

Femtosecond laser's enabling new length scale fabrications for rapid post irradiation examination of materials

PROJECT NUMBER:
22A1059-027FP

TOTAL APPROVED AMOUNT:
$978,000 over 3 years

PRINCIPAL INVESTIGATOR:
Stephanie Pitts

CO-INVESTIGATORS:
Brennan Harris, INL
David Frazer, INL
Peter Hosemann,
University of California, Berkeley

Connecting the yield stress behavior of nuclear-relevant materials to the gauge size and sample microstructural characteristics enables the reduction of mechanical testing sample sizes.

Mechanical testing campaigns are required to qualify materials for advanced reactor conditions, yet economical and safety limitations restrict the number of standardized mechanical tests that can be performed. Reducing the sample size is one approach to address these challenges and to accelerate testing. Previous research, however, has shown that smaller mechanical test samples produce higher yield and ultimate stress values compared to values measured from standard sample sizes: the "smaller is stronger" effect. Specimens used in accelerated material testing campaigns must reflect bulk material performance to enable engineering scale material property measurement. The objective of this research project was to determine whether engineering scale mechanical behavior—the yield stress—could be measured with micro-tensile test samples smaller than traditional standard testing geometries. The relationship between yield stress and sample size was explored with two different nuclear-relevant structural materials: Zircaloy-4 and tungsten. Mechanical testing of both metals demonstrated decreasing yield stress values with increasing sample gauge size across three different sizes. Yield stress values from the largest gauge size, 100 µm x 80 µm x 233 µm and fabricated with a femtosecond laser ablation system, approach bulk material yield stress values reported in published literature. Preliminary analysis of the tungsten samples indicates the yield stress value depends on the grain characteristics within the gauge section in addition to the gauge size. Accompanying modeling efforts, including response surface generation and crystal plasticity approaches, further demonstrated that the size of the sample gauge section alone cannot explain the change in yield stress values. Further study of the role of grain boundaries as strength-determining features could elucidate the connection between grain boundary characteristics and yield stress. This project demonstrated the use of a femtosecond laser ablation system to fabricate reduced size tensile specimens while retaining the ability to measure bulk material performance. The capability demonstrated here has the potential to accelerate mechanical testing campaigns for nuclear-relevant materials.

TALENT PIPELINE:

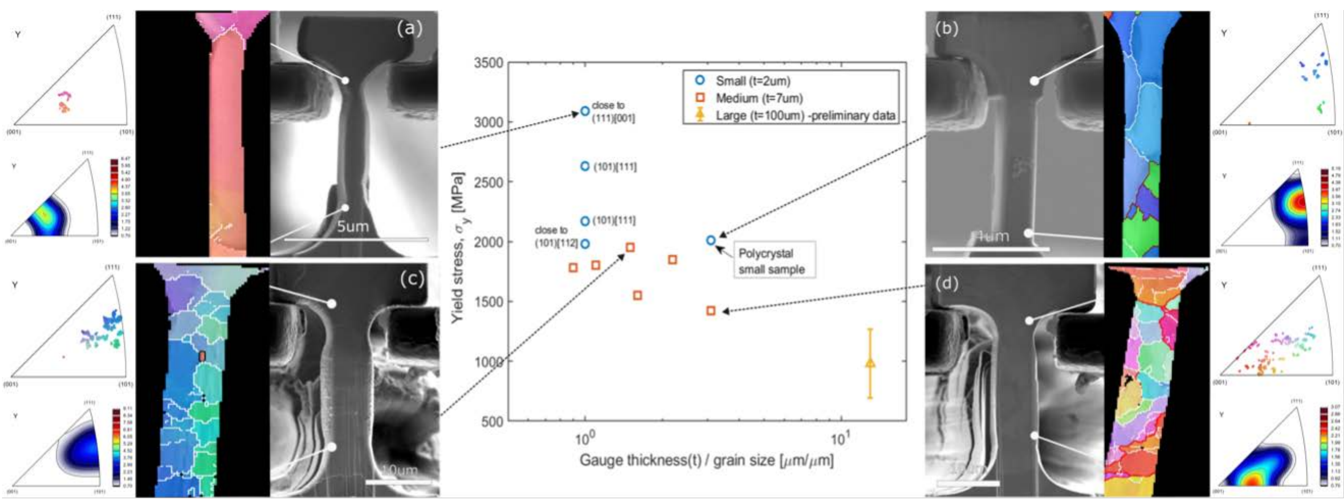
- Sebastian Lam, student at University of California, Berkeley
- Tanuj Gupta, student at Clemson University
- Tzu-Yi Chang, student at Oregon State University

PUBLICATION:

- Lam, S., Frazer, D., Cappia, F., Nelson, M., Samuha, S., Pitts, S., Harris, B., and P. Hosemann, "Length scale effects of micro- and meso-scale tensile tests of unirradiated and irradiated Zircaloy-4 cladding," Journal of Nuclear Materials, 155496, October 2024.

INTELLECTUAL PROPERTY:

- Harris, B., and S. Pitts, "MURMUR: Mesh Computing Remote Automatic Workflow," Open-Source Software Release, August 2024. Available at https://github.com/idaholab/MURMUR



Results for the yield stress values of tungsten micro-tensile specimens from three sample gauge sizes: small (2 µm x 2 µm x 7 µm), medium (7 µm x 7 µm x 18 µm), and large (100 µm x 80 µm x 233 µm). The small samples include both single crystals (a) and a polycrystalline (b) microstructure. The medium samples are all polycrystalline (c and d).

Multiphysics inversion for quantified nuclear material and system

PROJECT NUMBER:
22A1059-042FP

TOTAL APPROVED AMOUNT:
$1,277,000 over 3 years

PRINCIPAL INVESTIGATOR:
Lynn Munday

CO-INVESTIGATORS:
Dewen Yushu, INL
Kyle Paaren, INL
Oana Marin, INL
Yifeng Che, INL
Zachary Prince, INL
Murthy Guddati, Symulation, LLC

The multiphysics object-oriented simulation environment optimization module provides gradient based algorithms for model calibration and shape optimization.

Simulation reliability is critical to accelerate development of new nuclear technologies. The multiphysics object-oriented simulation environment (MOOSE) estimates safety and sustainability of many processes and systems related to nuclear energy. MOOSE simulation reliability is in part limited by the model parameters describing nuclear fuels. These parameters are difficult or expensive to measure experimentally. Inverse optimization provides a mathematical framework where parameter estimation is fit to system level observations. In this work, we developed a new optimization module in MOOSE to solve inverse problems using gradient based methods with a focus on parameter inversion for nuclear fuels. The software development focused on simplifying the implementation of new optimalization algorithms by using MOOSE features such as automatic differentiation and parsed function input of the objective function with an automatic adjoint formulation. The MOOSE optimization module provides researchers with a gradient based optimization platform to rapidly prototype and explore new optimizations algorithms tailored to their complex multiphysics problems.

During this work, we developed a protocol to use the MOOSE optimization module to solve nuclear fuel related inverse optimization problems where domain experts worked with optimization analysts to combine finite element modeling with sensitivity analysis to produce useful model parameterizations. Two nuclear fuel case studies were used to demonstrate the optimization protocol. The first study focused on high-power research reactor plate fuel to determine residual stress and power density of the low enriched uranium fuel. A coarse mesh of the finite element model used in the predictive simulations was used to parameterize the spatially varying residual stresses and power density. In the second study, we performed sensitivity analysis and parameterization of a metallic fuel model used to predict cladding deformation observed in post irradiation examination of Experimental Breeder Reactor-II pins.

TALENT PIPELINE:

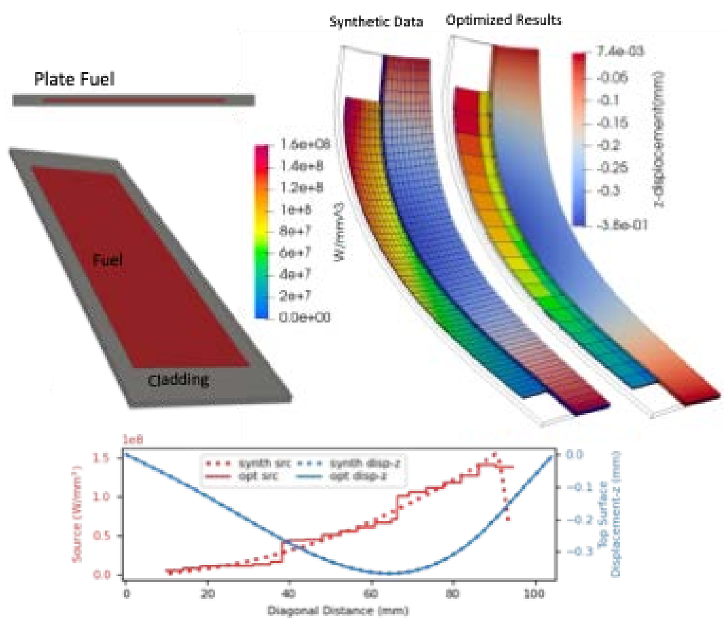
- Abd Che, *Russel L. Heath distinguished postdoctoral fellow at INL*

PRESENTATIONS AND PUBLICATIONS:

- Prince, Zachary M., Lynn Munday, Dewen Yushu, Max Nezdyur, and Murthy Guddati, "MOOSE Optimization Module: Physics-constrained optimization," SoftwareX 26 (2024): 101754.
- Elmeliegy, Abdelrahan, Matthew Urban, Lynn Munday, Murthy Guddati, "3D Multi-plane Multi-resolution Shear Wave Elastography," IEEE Ultrasonics, Ferroelectrics, and Frequency Control Joint Symposium (2024).
- Munday, Lynn, Zachary Prince, Dewen Yushu, Murthy Guddati, "MOOSE Optimization Module Overview: Application to Residual Stress Inversion in Nuclear Fuel Plates," US National Congress on Computational Mechanics 2023.

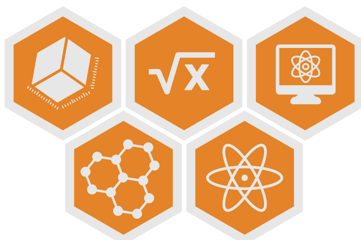
INTELLECTUAL PROPERTY:

- MOOSE Optimization module: https://mooseframework.inl.gov/modules/optimization/index.html



Thermomechanical inversion problem for plate fuel power density. An unknown spatially distributed heat source produced by the power distribution in the fuel was parameterized using the out-of-plane displacement data from the cladding produced from a simulation. In this example, the MOOSE optimization module performs coupled multiphysics inversion where measurements from one physics (mechanics) are used to invert for a material parameter from a different physics (heat conduction).

Accelerating deployment of nuclear fuels through reduced-order thermo-physical property models and machine learning



PROJECT NUMBER:
22A1059-084FP

TOTAL APPROVED AMOUNT:
$1,460,000 over 3 years

PRINCIPAL INVESTIGATOR:
Tsvetoslav Pavlov

CO-INVESTIGATORS:
Aysenur Toptan, INL
Congjian Wang, INL
Marat Khafizov, The Ohio State University

A computational framework leveraging solid state physics and machine learning interprets, predicts, and reduces the number of thermo-physical property experiments performed on nuclear fuels.

This project aimed to accelerate the deployment of novel nuclear materials by developing a novel data analysis tool that consists of computationally efficient solid state physics models and machine learning algorithms. The models describe thermal conductivity as a function of temperature and irradiation, leading to defect production, compositional changes, etc. Understanding and predicting the evolution of these material properties during reactor operation is critical for establishing fuel centerline temperature, fission gas release, and fuel rod pressure margins. These parameters are critical for licensing new nuclear fuels and enabling the design of new reactor concepts. The new tools will enable users to interpret measured thermo-physical property datasets in real time. In addition, the Platform for Optimal Experiment Management (POEM) was developed using machine learning algorithms. The computational efficiency of the new reduced-order models allows the user to employ POEM and train the physics-based models on available datasets and extrapolate to design targeted experiments. Ultimately, our results reduce the number of experiments required for fuel qualification. The long-term target is reducing the number of thermo-physical property characterization experiments by a factor of ten or more. The current project developed or applied: 1) computationally efficient solid state physics models, 2) machine learning platform POEM, 3) fuel performance methods, and 4) access to state-of-the-art thermal property characterization equipment at INL's Materials and Fuels Complex and irradiated nuclear fuel datasets. The methodologies were tested and validated against unirradiated metallic alpha uranium, unirradiated uranium dioxide and irradiated mixed oxide fuels at various burnups.

TALENT PIPELINE:

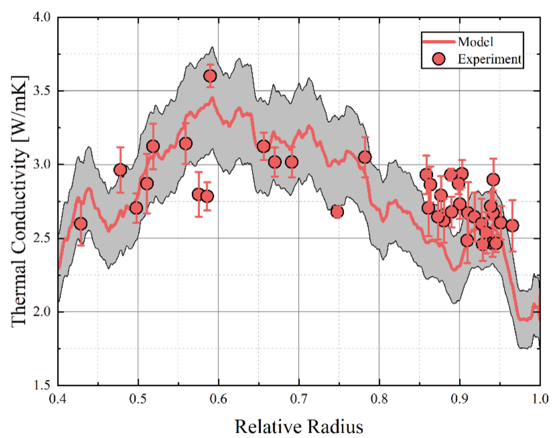
- Joshua Ferrigno, student at The Ohio State University
- Mutaz Alshannaq, student at The Ohio State University

PUBLICATION:

- Ferrigno, Joshua, Tsvetoslav Pavlov, Narayan Poudel, Daniele Salvato, Chuting Tsai, Brian Merritt, Alex Hansen, Troy Munro, Fabiola Cappia, and Marat Khafizov, "Analysis of radially resolved thermal conductivity in high burnup mixed oxide fuel and comparison to thermal conductivity correlations implemented in fuel performance codes," Journal of Nuclear Materials 596 (2024): 155090.

INTELLECTUAL PROPERTY:

- Open source, C. Wang, T. R. Pavlov, P. C. Simon, J. M. Ferrigno, "Platform for Optimal Experiment Management (POEM)," July 2024.



Estimated radial thermal conductivity of an irradiated mixed oxide fuel sample with a burnup of 6% fissions per initial metal atom. The Lucuta-Inoue approach was applied to model the evolution of thermal conductivity as a function of radius at room temperature. The model incorporates radial measurement inputs of porosity and burnup, along with model calculations of the average temperature profile and the linear stoichiometry approximation. The shaded area indicates a 10% error margin.

Promoting sparse sensing and sparse learning for nuclear digital twins

PROJECT NUMBER:
22A1059-091FP

TOTAL APPROVED AMOUNT:
$1,971,000 over 3 years

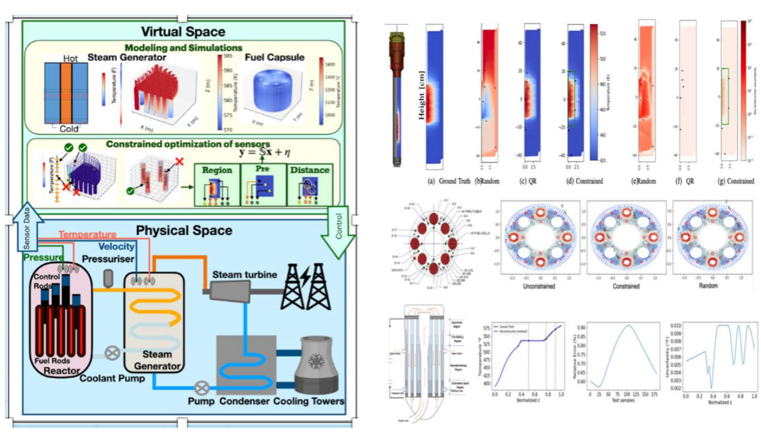
PRINCIPAL INVESTIGATOR:
Mohammad Abdo

CO-INVESTIGATORS:
Joshua Cogliati, INL
Pattrick Calderoni, INL
Krithika Manohar, University of Washington
Steven Brunton, University of Washington

COLLABORATOR:
University of Washington

Optimal sensor placement for reactor fields reconstruction enables communication among a network of nuclear digital twins.

Nuclear power plants require continuous monitoring of various systems, structures, and components to ensure safe and efficient operations. Accurate reconstruction of fields of interest (e.g., temperature, pressure, velocity, etc.) from sensor measurements is crucial to establish a two-way communication between physical experiments and models. However, deploying extensive sensor arrays in nuclear reactors is infeasible due to challenging operating conditions and inherent spatial limitations. In this work, we developed a data-driven technique that incorporates constraints into an optimization framework for sensor placement. The primary objective was to minimize reconstruction errors under noisy sensor measurements. To validate our methodology, we applied the algorithm to the out-of-pile Testing and Instrumentation Transient Water Irradiation System prototype capsule. This capsule is electrically heated to emulate the neutronic effect of the nuclear fuel. The Transient Water Irradiation System prototype that will eventually be inserted in the Transient Reactor Test Facility at INL, serves as a practical demonstration. Optimized constrained sensors reconstruct the field of interest within a tristructural isotropic fuel irradiation experiment, a lumped parameter model of a nuclear fuel test rod, and a steam generator. The resulting sensor-based temperature reconstruction within out-of-pile Testing and Instrumentation Transient Water Irradiation System demonstrates minimized reconstruction errors, provides probabilistic bounds for noise-induced uncertainty, and establishes a foundation for communication between the digital twin and the experimental facility.



The left side of the figure illustrates the interaction between the physical world and its digital counterpart. The right side highlights three case studies demonstrating the application of optimal sensor placement. First, the electrically heated out-of-pile Testing and Instrumentation Transient Water Irradiation System prototype replicates the coolant system of a fuel pin. Second, a graphite holder for tristructural isotropic annular fuel, where over 20 bores were drilled to accommodate thermocouples for temperature monitoring and failure detection, was optimized by algorithms that reduced the required sensors to just three, achieving accurate three-dimensional temperature field reconstruction. Finally, a simplified steam generator was instrumented to reconstruct fields such as velocity, temperature, and heat flux, all with a maximum error of less than 2%.

TALENT PIPELINE:

- Haeseong Kim, student at Massachusetts Institute of Technology
- Niharika Karnik, student at University of Washington

PRESENTATIONS AND PUBLICATIONS:

- Karnik, N., Abdo, M.G., Estrada-Perez, C.E., Yoo, J.S., Cogliati, J.J., Skifton, R.S., Calderoni, P., Brunton, S.L. and Manohar, K., "Constrained optimization of sensor placement for nuclear digital twins," IEEE Sensors Journal, Volume 24, Issue 9, 2024.
- Karnik, N., Wang, C., Bhowmik, P. K., Cogliati, J. J., Balderrama Prieto, S. A., Xing, C., Klishin A.A., Skifton, R., Moussaoui, M., Folsom, C.P., Palmer, J.J., Sabharwall, P., Manohar, K., and Abdo, M. G., "Leveraging Optimal Sparse Sensor Placement to Aggregate a Network of Digital Twins for Nuclear Subsystems," Energies (19961073), 17(13), 2024.
- Kim, H., Cetiner, S., and Bucci, M., "Thermal fluids field reconstruction with Bayesian inference for forced convection system," Proceedings of 13th Nuclear Plant Instrumentation, Control and Human-Machine Interface Technologies, American Nuclear Society, NPIC and HMIT 2023 (pp. 639-648), 2023.
- Kim, H., Cetiner, M., and Bucci, M., "Inverse Problem Approach for Estimating Operating Conditions and Uncertainty Quantification in Actual Forced Convection System," International Congress on Advances in Nuclear Power Plants (ICAPP), 2024.

INTELLECTUAL PROPERTY:

- Pysensors - https://github.com/dynamicslab/pysensors
- RAVEN Sparse Sensing Postprocessor https://github.com/idaholab/raven/blob/devel/ravenframework/Models/PostProcessors/SparseSensing.py

AWARDS

- 1st place best poster award, 2024 LDRD projects at INL
- Best poster award in the mechanical engineering department at University of Washington

Artificial intelligence enhanced advanced post irradiation examination

PROJECT NUMBER:
22A1059-094FP

TOTAL APPROVED AMOUNT:
$1,423,000 over 3 years

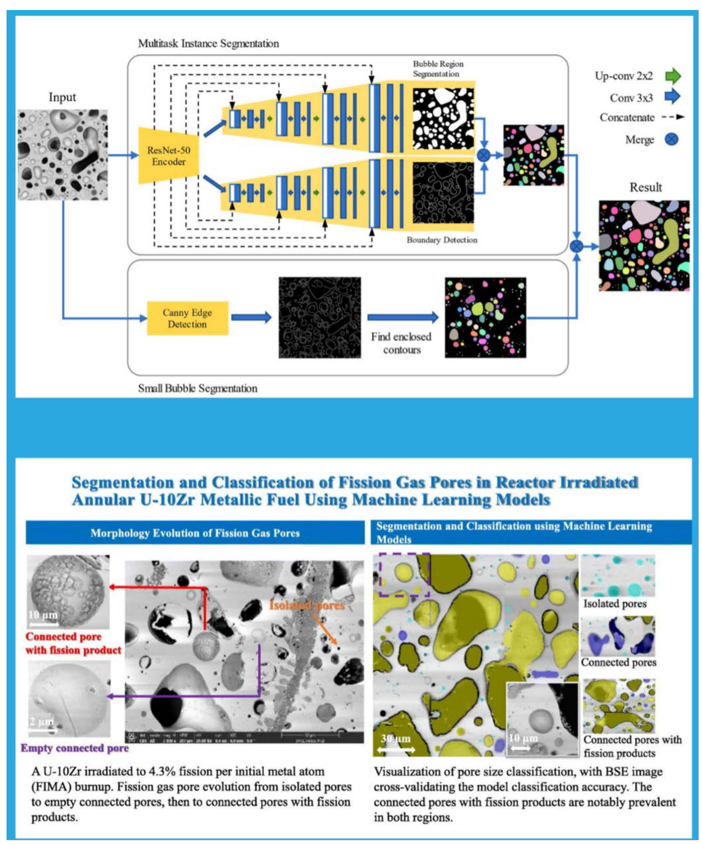
PRINCIPAL INVESTIGATOR:
Tiankai Yao

CO-INVESTIGATORS:
Cynthia Adkins, INL
Daniel Murray, INL
Larry Aagesen, INL
Lu Cai, INL
Luca Capriotti, INL
Min Xian, University of Idaho

COLLABORATOR:
University of Idaho

Artificial intelligence models reduce data analysis and quantification time and quantify fuel microstructure and changes caused by neutron irradiation.

Uranium zirconium metallic nuclear fuel is the leading candidate for next generation sodium-cooled fast reactors in the US. Material characterization involves analyzing the physical and chemical properties of various materials and is crucial for understanding the fuel performance and providing supports for fuel qualification. Quantitively analysis on material characterization data accelerates the research cycle and provide unprecedented, quantified insights into the evolution of material microstructures during irradiation process, such as fission gas pore, and lanthanide and zirconium distribution. In this project, the team collected first-of-kind post irradiation examination data on types of irradiated uranium zirconium fuel and developed several artificial intelligence models for fission gas bubble segmentation and classification. Specifically, several fission gas bubble detections related deep learning models were developed, and the fission gas bubbles were segmented and classified into different categories to better reveal the connection between fission gas bubbles and lanthanide liquid like movement in irradiated metallic fuel. Those models quantified microstructure features and increased data analysis efficiency. The outcomes from the research better revealed irradiation effects in reactor irradiated nuclear fuels.



Deep learning model for fission gas pore detection and the model's quantitative analysis of fission gas pore on irradiated annular uranium zirconium (U-10Zr) metallic fuel.

TALENT PIPELINE:

- Arnold Pradhan, student at University of Idaho
- Daniele Salvato, postdoc at INL, converted to staff
- Fei Xu, postdoc at INL, converted to staff
- Haotian Wang, student at University of Idaho
- Liang Zhao, student at University of North Carolina, hired as postdoc at INL
- Shoukun Sun, student at University of Idaho
- Yalei Tang, student at University of Nebraska

PRESENTATIONS AND PUBLICATIONS:

- Wang, Y., B.D. Miller, J.M. Harp, M.N. Bachhav, L. Capriotti, and T. Yao, "Advanced Characterization of Fuel Cladding Chemical Interaction between U-10Zr Fuel and HT9 Cladding Tested in Fast Flux Test Facility," Microscopy and Microanalysis 28, no. S1 (2022): 2106-2107.
- T. Yao, X. Liu, Y. Wang, F. Teng, D.J. Murray, M. Meyer, M.T. Benson, L. Capriotti, "Transmission Electron Microscopy based Characterization of a U-20Pu-10Zr Fuel Irradiated in Experimental Breeder Reactor-II," Journal of Nuclear Materials, 2022, 568, 153846.
- L. Cai, F. Xu, F. G. Di Lemma, et al., "Understanding fission gas bubble distribution, lanthanide transportation, and thermal conductivity degradation in neutron-irradiated α-U using machine learning," Materials Characterization, 2022, 184, 111657.
- D. Salvato, X. Liu, D.J. Murray, K.M. Paaren, F. Xu, T. Pavlov, M.T. Benson, L. Capriotti, T. Yao, "Transmission electron microscopy study of a high burnup U-10Zr metallic fuel," Journal of Nuclear Materials, 2022, 570: p.153963.
- Yao, T., M. Bachhav, F. Di Lemma, F. Xu, F. Teng, D.J. Murray, M.T. Benson, and L. Capriotti, "The advanced characterization, post-irradiation examination, and materials informatics for the development of ultra high-burnup annular U-10Zr metallic fuel," Frontiers in Nuclear Engineering 1 (2023): 1050262.
- Sun, S., F. Xu, L. Cai, D. Salvato, L. Capriotti, M. Xian, and T. Yao, "An efficient instance segmentation approach for studying fission gas bubbles in irradiated metallic nuclear fuel," Scientific Reports 13, no. 1 (2023): 22275.
- Xu, F., L. Cai, D. Salvato, L. Capriotti, and T. Yao, "Advanced characterization-informed machine learning framework and quantitative insight to irradiated annular U-10Zr metallic fuels," Scientific Reports 13, no. 1 (2023): 10616.
- Wang, H., F.ei Xu, L. Cai, D. Salvato, F. Di Lemma, L. Capriotti, T. Yao, and M. Xian, "A fine pore-preserved deep neural network for porosity analytics of a high burnup U-10Zr metallic fuel," Scientific Reports 13, no. 1 (2023): 22274.
- Sun, S., M. Xian, F. Xu, L. Capriotti, and T. Yao, "Cfr-icl: Cascade-forward refinement with iterative click loss for interactive image segmentation," Proceedings of the AAAI Conference on Artificial Intelligence, vol. 38, no. 5, pp. 5017-5024. 2024.
- Salvato, D., K.M. Paaren, J.A. Hirschhorn, L.K. Aagesen, F. Xu, F. Di Lemma, L. Capriotti, and T. Yao, "The effect of temperature and burnup on U-10Zr metallic fuel chemical interaction with HT-9: A SEM-EDS study," Journal of Nuclear Materials 591 (2024): 154928.
- Tang, Y., F. Xu, S. Sun, D. Salvato, F. Di Lemma, M. Xian, D.J. Murray, C. Judge, L. Capriotti, and T. Yao, "Segmentation and classification of fission gas pores in reactor irradiated annular U10Zr metallic fuel using machine learning models," Materials Characterization (2024): 114061.
- A. Pradhan, F. Xu, D. Salvato, I. Charit, C. Judge, L. Capriotti, T. Yao, "Characterization of Fuel Cladding Chemical Interaction on a High Burnup U-10Zr Metallic Fuel via Electron Energy Loss Spectroscopy Enhanced by Machine Learning," Material Characterization, 2024: 114524.
- Xu, F., Cai, L., Salvato, D. et al., "A Fine Pore-preserved Deep Neural Network for Porosity Analytics of a High Burnup U-10Zr Metallic Fuel," The Minerals, Metals & Materials Society (TMS) 2024, Orlando.
- Tang, Y., Xu, F., Yao, T., "Quantitative Insight to Fission Gas Pores Distribution in Irradiated Annular U-10Zr Metallic Fuel Using Machine Learning," TMS 2024, Orlando.
- A. Pradhan, F. Xu, L. Cai, D. Salvato, T. Yao, "Electron Energy Loss Spectroscopy (EELS) Characterization of Fuel Cladding Chemical Interaction (FCCI) region in U-Zr Metallic Fuel cladded with HT-9," TMS 2024, Orlando.
- F. Di Lemma, S Vajayan, D. Salvato, K. Wright, L. Hawkins, K. Bawane, L. Capriotti, T. Yao, "In-situ TEM Studies of Microstructural and Phase Evolution in Metallic Fuel Alloys," Materials in Nuclear Energy Systems (MiNES 2023).
- C. Adkins, D. Salvato, T. Yao, "A Study in the Thermal Transport Properties of Sodium-Bonded U-Zr Metallic Fuels for Fast Reactors," Materials in Nuclear Energy Systems (MiNES 2023).
- D. Salvato, A. Pradhan, F. Xu, K. Paaren, F.Teng, F. Di Lemma, L. Capriotti, T. Yao, "Transmission Electron Microscopy Study of the Fuel Cladding Chemical Interaction Layer Forming in a U-10Zr Fuel Sample Irradiated to High Burnup," Materials in Nuclear Energy Systems (MiNES 2023).

Microstructurally driven framework for optimization of in-core materials

PROJECT NUMBER:
22A1059-109FP

TOTAL APPROVED AMOUNT:
$1,815,000 over 3 years

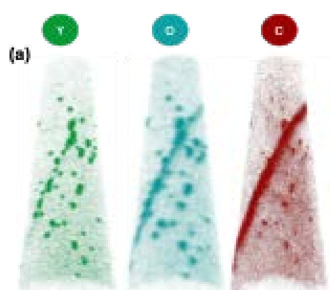
PRINCIPAL INVESTIGATOR:
Mukesh Bachhav

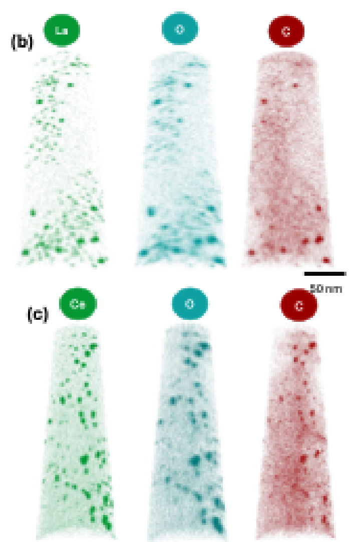
CO-INVESTIGATORS:
Douglas Porter, INL
Tiankai Yao, INL
Sohail Shah, INL
Xinchang Zhang, INL
Frank Garner, Texas A&M University
Lin Shao, Texas A&M University

COLLABORATOR:
Clemson University
Texas A&M University

Advanced manufacturing and machine learning-assisted molecular dynamic modeling accelerates structural alloy development for advanced reactors.

Accelerating early-to-mid stage development of critical in-core materials requires methodologies that include alloy optimization to discover performance issues to reduce near-term risks to advanced reactor deployment. The current fast reactor concepts require cladding materials to sustain low and predictable creep and swelling rates to 250–500 displacement per atom with service life extending to 15–20 years, well beyond the existing irradiation database. The inherent properties of steel like swelling resistance, low radiation damage accumulation, and good thermal conductivity are further enhanced by the presence of oxide nanoparticles finely distributed in the microstructure. In this project, iron-chromium-molybdenum alloys were fabricated by suspending oxide nanoparticles of yttrium, lanthanum, and cerium within the microstructure. The microstructure, microhardness, and tensile properties of the fabricated variants were investigated. Additionally, the team developed a methodology that relies on mechanism-informed machine learning models, rapid ion irradiation and creep testing techniques, and advanced characterization coupled with automated image analysis that allow reactor developers to quickly understand the complex linkage between alloy composition, thermomechanical processing, the resulting microstructure, and swelling and creep behavior. A novel methodology based on advanced manufacturing was developed and demonstrated to enable rapid development of future in-core materials.





Atom probe tomography maps of (a) yttrium (Y), (b) lanthanum (La) and (c) cerium (Ce) in green along with other elements, oxygen (O) and carbon (C) seen within oxide particles.

TALENT PIPELINE:

- Carel Ziminsky, student at Clemson University
- Jack Eggemeyer, student at Clemson University
- Safqut Sanwar, student at Clemson University
- Sohail Shah, postdoc at INL

PUBLICATIONS:

- KS Mao, H Wang, HJ Qu, KH Yano, PD Edmondson, C Sun, JP Wharry, "Probing the Damage Recovery Mechanism in Irradiated Stainless Steels Using In-Situ Microcantilever Bending Test," Frontiers in Materials 9, 823192.
- X Zhang, Q Wang, JJ Kane, JF Rufner, C Sun, "Graded microstructure and mechanical properties of spark plasma sintered Fe-Cr alloys," Journal of Alloys and Compounds 967, 171448.
- X Zhang, L Wang, F Liou, Y Ren, C Sun, "Microstructure and Residual Stress in Functionally Graded 316L Stainless Steel/Inconel 625 Alloys Fabricated by Direct Energy Deposition," Jom 75 (12), 5066-5078.
- M Pena, Z Hu, Y Li, C Sun, SA Maloy, FA Garner, L Shao, "Formation and dissolution of carbides and precipitates in self-ion irradiated HT9 alloy," Journal of Nuclear Materials 588, 154819.

Novel in situ measurements enabled by advanced modeling and data analytics to accelerate nuclear technology deployment via the Advanced Test Reactor

PROJECT NUMBER:
22A1059-134FP

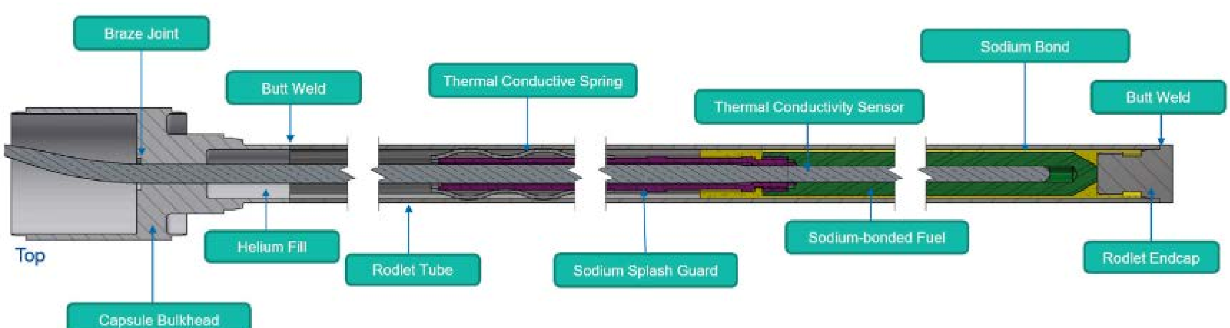
TOTAL APPROVED AMOUNT:
$3,901,777 over 3 years

PRINCIPAL INVESTIGATOR:
Austin Fleming

CO-INVESTIGATORS:
Colby Jensen, INL
Han Bao, INL
Nicolas Woolstenhulme, INL

First-of-kind capability enabled for in-core thermal conductivity measurements at the Advanced Test Reactor.

This project has established a transformational approach to conducting instrumented experiments in the Advanced Test Reactor through the use of new penetrations in the top head closure for providing instrumentation feed-throughs. A first-of-its-kind experiment design was created to utilize these penetrations for instrumented "drop-in experiments." This experiment employed an INL-developed thermal conductivity probe to measure changes in a specimen's thermal conductivity throughout neutron irradiation in the Advanced Test Reactor. The specimens in this irradiation experiment focus on metallic fuel composed of uranium with 10% by weight zirconium. The thermal conductivity of metallic fuel has never been measured in-core, and very limited data on irradiated properties are available in the literature. Three design variations of this metallic fuel were tested: a sodium-bonded design, a slotted fuel design, and an annular fuel design. Additionally, a reference specimen of stainless steel 304 is included in the experiment for measurement validation purposes. Due to delays with the reactor schedule, the irradiation of the experiment was not completed at the conclusion of the project. However, the work will continue through other DOE research programs.



Cross section view of the sodium-bonded metallic fuel specimen capsule for the irradiation experiment in the Advanced Test Reactor with the thermal conductivity sensor at the center of the specimen.

TALENT PIPELINE:

- Katelyne Wada, student at Boise State University
- Takanori Kajihara, postdoc at INL, converted to staff

PRESENTATIONS AND PUBLICATIONS:

- T. Kajihara, H. Bao, D. Chapman, S. Qin, A. Fleming, "A machine-learning-aided data recovery approach for predicting multi-material thermal behaviors in Advanced Test Reactor capsules," International journal of Heat and Mass Transfer, Volume 231, 12528, 2024.
- T. Kajihara, H. Bao, R. T. Sweet, A. Zabriskie, A. D. Fleming, "BISON-Machine-Learning Hybrid Approach for Predicting the Physics Behaviors in Advanced Test Reactor Capsules," The 14th International Topical Meeting on Nuclear Reactor Thermal-Hydraulics, Operation, and Safety, August 25–28, 2023.
- H. Bao, T. Kajihara, R. T. Sweet, A. Zabriskie, A. D. Fleming, "BISON-Machine-Learning Hybrid Approach for Predicting the Thermal Behaviors in Advanced Test Reactor Capsules," Advanced in Thermal Hydraulics (ATH 2024), 2024.
- C. Downey, N. Oldham, A. Fleming, D. Chapman, A. M. Cruz, K. Ellis, "Design of a first-of-a-kind instrumented Advanced Test Reactor irradiation capsule experiment for in situ thermal conductivity measurement of metallic fuel," Progress in Nuclear Energy, volume 175, 105325, 2024.
- Fleming, C. Jensen, N. Woolstenhulme, N. Oldham, "Experiment Design for the In-pile Measurement of U-10Zr," Thermal Conductivity, American Nuclear Society Annual Conference, Anaheim, CA, 2022.
- K. Wada, A. Fleming, C. Jensen, J. Eixenberger, B. Jaques, D. Estrada, "Transient multilayer analytical model of a line heat source probe for in-pile thermal conductivity measurements," International Journal of Thermophysics, 2023.
- K. Wada, A. Bateman, T Varghese, A Fleming, B. Jaques, D Estrada, "High temperature validation of a line heat source technique for in-pile thermal conductivity determination," International Journal of Heat and Mass Transfer, 2024.

A causal approach to model validation and calibration

PROJECT NUMBER:
23A1070-144FP

TOTAL APPROVED AMOUNT:
$910,000 over 2 years

PRINCIPAL INVESTIGATOR:
Diego Mandelli

CO-INVESTIGATORS:
Congjian Wang, INL
Mohammad Abdo, INL
Paolo Balestra, INL
Ron Gonzales, INL
Sunming Qin, INL

COLLABORATOR:
University of Michigan

Blending statistical conditional independence testing methods with regression models quantifies causal relations between observed data elements.

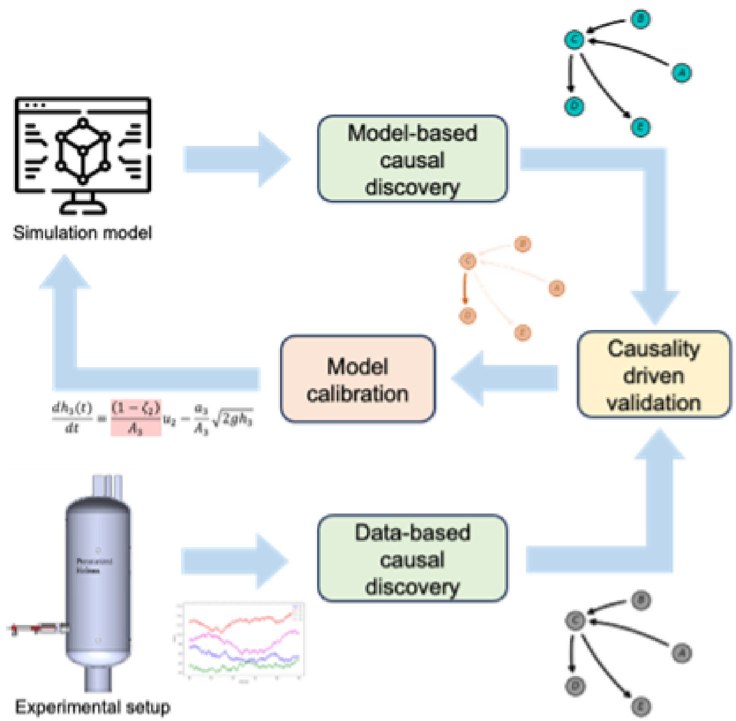
The nature of science and technology is the discovery and the control of the causal mechanisms behind observed phenomena. In the past few decades, machine learning and artificial intelligence have spread in almost all science and technology fields, but the employed methods have focused on fitting the data rather than understanding the data. This project brought a science-based mindset to artificial intelligence and machine learning methods with the goal of discovering and quantifying causal relationships between observed data elements. The developed methods embrace causality at their core by first identifying the physical relation among data elements and then by quantifying it in the form of structural causal models. The process of causal discovery was accomplished by blending statistical conditional independence testing methods to discover physical relations—the existence of a physical relation between variables implies that those variables are in fact dependent—with regression models to quantify the amount of these causal relations. Once causal relations are inferred, deviations of system behavior can be quantified in terms of structural causal models—different strength of causal relations or new or missing causal relations. By doing this, the source(s) of such deviation can be identified, and possible counteractions can be established. Note that our causal approaches are more interpretable since any root cause analysis must match our physical understanding and align with expert domain knowledge. The application of our methods is the validation of simulation models. Here, the process of comparing simulated and experimental setups is framed in terms of generation and comparison of structural causal models. A major challenge of current causal discovery approaches is their robustness to different operational conditions and the reliability of obtained results. Unlike current causal discovery approaches, the development and refinement of our methods have been tested through many operational conditions (e.g., non-linear casual behaviors, presence of noise in the observed data) using both analytical and experimental datasets.

TALENT PIPELINE:

- Ron Gonzales, postdoc at INL

PRESENTATIONS:

- D. Mandelli, R. Gonzales, C. Wang, M. Abdo, Z. Welker, P. Balestra, S. Qin, V. Petrov, "A Causal Approach to Model Validation and Calibration," Proceedings of the ASME 2023 – International Mechanical Engineering Congress and Exposition IMECE2023 (2023).
- R. Gonzales, D. Mandelli C. Wang, M. Abdo, Z. Welker, P. Balestra, S. Qin, V. Petrov, "Verification, Validation, And Calibration Through a Causal Lens," ASME Verification, Validation, and Uncertainty Quantification Symposium (2024).



Causal approach to model validation: causal discovery methods applied to simulation models and observed experimental data are designed to generate structural causal models which are then compared to assist analysts on the identification of the equation parameters of the simulation model that needs to be modified (calibration).

Production of high specific activity germanium-71 through photonuclear reactions and nuclear recoil separation

PROJECT NUMBER:
23P1079-014FP

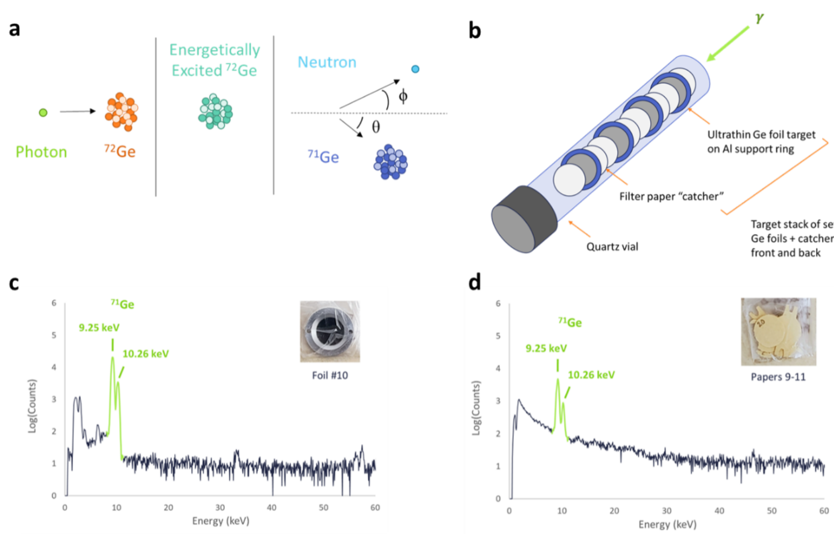
TOTAL APPROVED AMOUNT:
$123,400 over 1 year

PRINCIPAL INVESTIGATOR:
Emily Paige Abel

CO-INVESTIGATORS:
Nick Erfurth, INL
Mason Jaussi, Idaho State University

Auger electron emitting radionuclides can be potent therapeutic agents against metastatic cancer with fewer negative side effects as compared to other therapies.

Targeted internal radiotherapy is a treatment technique for metastatic cancers that uses ionizing radiation such as alpha, beta, and Auger electron emissions to damage cancer cells. Recently, Auger electron emitting radionuclides have gained attention for their dense energy deposition over a short range. This characteristic means they have potential for effective therapy and minimal side effects compared to other radiotherapies. However, common production routes such as thermal neutron capture in a reactor or the emission of a neutron via a photonuclear reaction rely on irradiating a target material of the same element as the product nucleus. This results in a product that has unacceptably low specific activity to be used for medical applications. Traditionally, time-consuming and expensive mass separations are needed to isotopically purify the Auger electron product following these production methods. A new method of in-line separation using nuclear recoil and ultrathin target foils has been used to produce high specific activity germanium-71 from a natural germanium target via a photonuclear reaction. Following the absorption of a photon and subsequent neutron emission, the product nucleus is imparted with a small amount of kinetic energy due to nuclear recoil. With micrometer-scale target foils, the product nucleus has enough energy to escape from the thin foil a fraction of the time. Scientists at INL and Idaho State University conducted experiments at the Idaho Accelerator Center to test this novel separation method and measure the production separation efficiency for germanium-71 from natural germanium-71 foils. While the separation efficiency was relatively modest for this experiment, the results have been compared to nuclear recoil models, which will help to inform future experiments as this method is further developed.



Nuclear recoil separation for isotope production: a) Cartoon of photonuclear production of germanium-71 (^{71}Ge) and nuclear recoil following the nuclear reaction, b) Drawing of target used for irradiation of natural germanium foils for the production and separation of ^{71}Ge, c) Gamma spectrum of a natural germanium foil irradiated with photons showing characteristic x-ray emissions from ^{71}Ge and demonstrating production of desired product, d) Gamma spectrum of "catcher" material used to capture ^{71}Ge recoiling from target foils showing characteristic x-rays from ^{71}Ge and demonstrating nuclear recoil separation.

Development of numerical stand-ins for design of autonomous reactor control systems in the Multiphysics Object-Oriented Simulation Environment

PROJECT NUMBER:
23P1079-019FP

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PRINCIPAL INVESTIGATOR:
Stefano Terlizzi

CO-INVESTIGATORS:
Sebastian Schunert, INL
Brendan Kochunas, University of Michigan

Modeling instrumentation and control systems with full core multiphysics simulations advances autonomous microreactors.

Researchers demonstrated for the first time the ability to model instrumentation and control systems within a MOOSE based multiphysics full core simulation of a realistic heat pipe-cooled microreactor. This was achieved by developing new functionalities within MOOSE to simulate the transient and steady-state responses of sensors in a standardized and efficient manner. The sensor response was characterized using six parameters: (1) signal-to-noise ratio, (2) noise standard deviation, (3) signal delay, (4) signal drift, (5) sensor efficiency, and (6) sensor uncertainty. This lumped parameter approach provides high flexibility in defining responses for different sensor types, including thermocouples and neutron counters. These new capabilities were integrated with existing MOOSE-based controller logic, demonstrating the combined use of sensors and controls in MOOSE. This represents the first instance of modeling instrumentation and control systems interacting with a full core multiphysics simulation of a realistic heat pipe-cooled microreactor, proving the feasibility of using MOOSE for such applications. This research established a strategic partnership with Dr. Kochunas' laboratory at the University of Michigan to advance automated controls for microreactors in line with the objective of designing fully autonomous fission batteries.

TALENT PIPELINE:

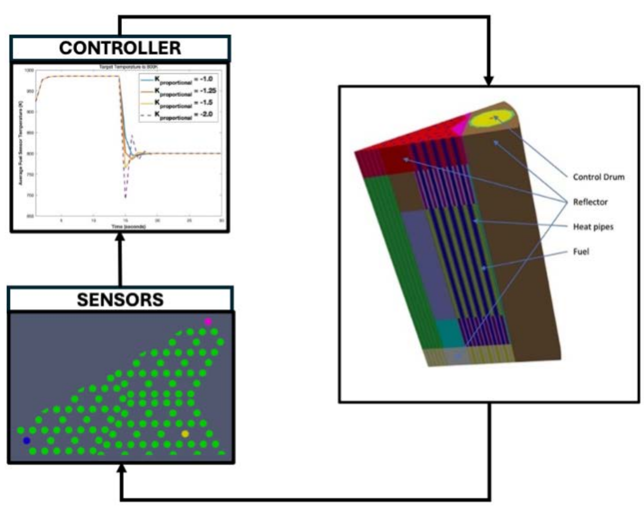
- Farhana Farha, student at the University of Michigan

PUBLICATION:

- F. Farha, J. Hanophy, Brendan Kochunas, Sebastian Schunert, and Stefano Terlizzi, "Proof-of-Concept of Sensor Modeling in MOOSE for the Design of Autonomous Nuclear Reactor Control," Proceedings of the 2024 ANS Winter Conference and Expo, Orlando, Florida, November 17–21, 2024.

INTELLECTUAL PROPERTY:

- General Sensor Postprocessor, https://github.com/idaholab/moose/pull/26870



Schematics of MOOSE-based integration between nuclear microreactor full core problem, sensors, and controllers.

Liquid fuel testing capability at the Advanced Test Reactor

PROJECT NUMBER:
23P1082-012FP

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PRINCIPAL INVESTIGATOR:
Daniel Sluder

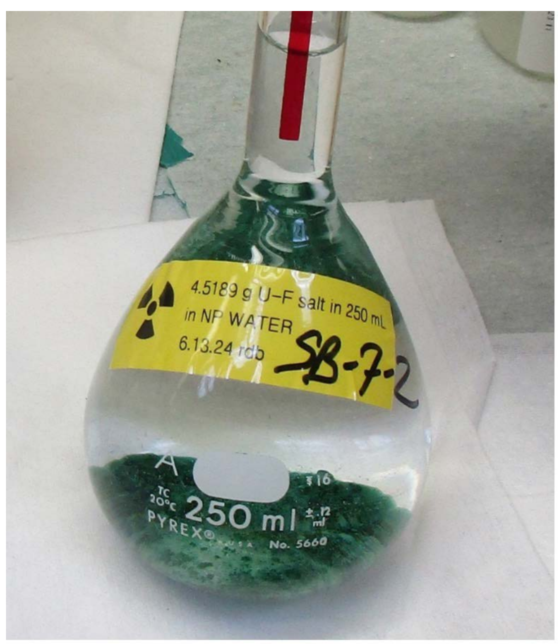
CO-INVESTIGATORS:
Igor Sarygin, INL
Jacob Westacott, INL
Nate Oldham, INL
R. Duane Ball, INL

This project paved the way for molten fuel irradiation experiments at the Advanced Test Reactor.

The Advanced Test Reactor is the world's most powerful research reactor. A primary mission of the Advanced Test Reactor is to support the next generation of nuclear reactors. This support requires irradiation of advanced fuels such molten salts and metal eutectics. Prior to this project, no hazard analysis had been completed for molten fuel experiments, and irradiation would have been prohibited under the Advanced Test Reactor safety analysis report. This project paved the way for molten fuel experiments within the Advanced Test Reactor safety basis by addressing three required items. The first item was an updated source term analysis. Molten fuels behave differently than other fuels during accident scenarios, so the Advanced Test Reactor safety basis was updated to account for radiological releases from molten fuel experiments. The second item was a quantification of the chemical response of the Advanced Test Reactor primary coolant system to an inadvertent release of molten fuel. This project developed a protocol to set fail-safe material limits on molten fuel experiments and determined the limits for two common fuel salts—sodium chloride-uranium chloride and uranium fluoride-sodium fluoride-potassium fluoride. The third item was creating guidance for the structural design of molten fuel experiments. Molten fuel experiments will need to operate at temperatures and pressures outside of standard design codes. It was necessary to recommend a structural design methodology for molten fuel experiments. With the combination of these three items completed, Advanced Test Reactor now has the capability to irradiate molten fuel samples.

PUBLICATION:

- D. K. Sluder, et al., "Safety Considerations for Advanced Material Irradiation at the Advanced Test Reactor," Trans. Am. Nucl. Soc., June 2024.



Titrations of uranium fluoride salt demonstrate insolubility and provide insights into molten salt experiment designs at the Advanced Test Reactor.

Hybridized discontinuous Galerkin methods for computational fluid dynamics

PROJECT NUMBER:
24A1083-012FP

TOTAL APPROVED AMOUNT:
$200,000 over 1 year

PRINCIPAL INVESTIGATOR:
Alexander Lindsay

Hybridizable discontinuous Galerkin methods complete high accuracy calculations in similar times as current finite volume methods.

Hybridizable discontinuous Galerkin methods hold promise for any applications with significant advection character, including thermal hydraulics in light water reactors and advanced reactor concepts and fluid models of plasmas in magnetic confinement fusion. Its features include natural upwinding, local element conservation, and extensibility to arbitrarily high-order accuracy. In this project, we implemented hybridizable discontinuous Galerkin methods in the MOOSE. We developed a first-of-its-kind automatic static condensation system in MOOSE's underlying finite element library libMesh, which can condense arbitrarily many internal variables. Finally, we developed the first preconditioner for hybridizable discontinuous Galerkin discretizations of the Navier-Stokes equations, which shows robust performance across a wide range of problem sizes and Reynolds numbers. This preconditioner yields solution times equivalent to the fastest developed for industry standard finite volume methods. Moreover, the arbitrarily high-order nature of hybridizable discontinuous Galerkin makes it a prime candidate for acceleration via graphical processing units. We believe these developments will hold significant importance for future DOE Nuclear Energy and Fusion Energy Science programs.

Streamlines for a lid driven cavity simulation with a Reynolds number of 10,000 discretization with the hybridizable discontinuous Galerkin method.

INTEGRATED FUEL CYCLE SOLUTIONS

We create effective and integrated fuel cycle solutions to sustain the commercial reactor fleet and enable expanded nuclear energy deployment. These efforts include developing new fuel cycle technologies and capabilities to enable advanced reactor systems, as well as technologies associated with management and disposition of existing and future radiological waste materials. Our activities contribute to the critical near-term high-assay low-enriched uranium needs for advanced reactors, and enable the broader and longer-term front-end fuel cycle needs. We are exploring backend fuel cycle technologies for advanced reactor demonstrations, including those that will occur on our site, and deployment over the next five to ten years. Specifically, the lab is focused on RD&D of recycling technologies to improve economics and reduce waste materials and proliferation risk by advancing safeguards and security through unique test bed capabilities.

CORE CAPABILITIES

Advanced Computer Science, Visualization, and Data

Applied Mathematics

Chemical Engineering

Decision Science and Analysis

Nuclear Engineering

Systems Engineering and Integration

EDNA CARDENAS

Advances in nuclear fuel cycle nonproliferation, safeguards, and security using an integrated data science approach

PROJECT NUMBER:
22A1059-073FP

TOTAL APPROVED AMOUNT:
$2,877,000 over 3 years

PRINCIPAL INVESTIGATOR:
Ashley Shields

CO-INVESTIGATORS:
Adam Pluth, INL
Cassandra Pate, INL
Eduardo Treviño, INL
Gustavo Reyes, INL
Jaren Brownlee, INL
Justin Cooper, INL
Katherine Jesse, INL
Mark Schanfein, INL
Ramedy Flores, INL

Digital twin provides near real-time, mixed reality visualization of centrifugal contactor operations.

This project established the foundations of a solvent extraction process digital twin for the Beartooth testbed. This provides an opportunity to gain a deeper understanding of the challenges and opportunities related to 1) the ingestion of high volume, high dimensionality timeseries data, 2) incorporation of in-line spectrographic sensors for analyte concentration monitoring, 3) timeseries analysis of spectral data with optical interference due to air entrainment, 4) the integration of MATLAB simulation, 5) developing visualization approaches for centrifugal contactor operations, specifically in the context of international safeguards. This was achieved by developing a digital twin based on a 30-stage centrifugal contactor solvent extraction testbed located at INL using the surrogate metals neodymium (extractable) and nickel (non-extractable) to develop capabilities relevant to the Purex process. This included the installation of ultraviolet and visible spectrum sensors for the purpose of online monitoring.

Data was uploaded to the DeepLynx data warehouse, an open source INL software, where it was then connected through backend software development for analysis and visualization. The analyses included a machine learning based anomaly detection, namely Isolation Forest along with traditional analytical chemistry methods for concentration prediction. Those traditional methods had previously been implemented manually via Microsoft Excel calculations, but our team translated those methods into a python code so that concentration predictions could be calculated in near real-time. Information is displayed in a web interface that includes data streams, modeling and simulation results, and data science results. This includes automated insights based on semantic constraints to alert the user to investigate further if unexpected conditions occur (e.g., unexpectedly high neodymium concentration). The data streams, modeling and simulation, and online monitoring analysis are also visualized in a mixed reality interface where the user can see that information within the context of a virtual version of operating contactors.

TALENT PIPELINE:

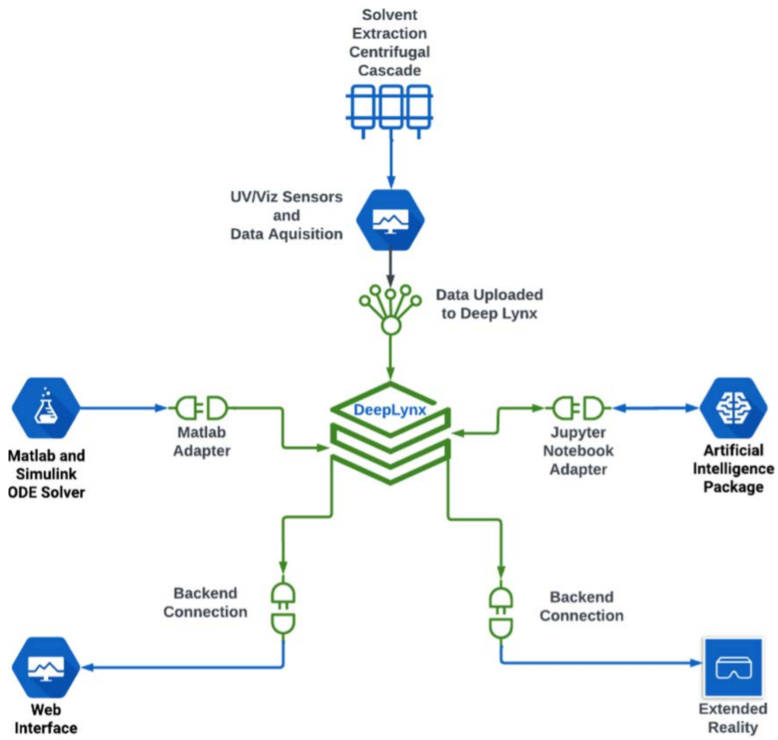
- Dawn Knoll, student at Brigham Young University–Idaho
- Dylan Kuncheria, student at University of Wisconsin, Madison, converted to staff
- Eduardo Treviño, student at University of Texas, San Antonio, converted to staff
- Hannah Warren, student at Northern Arizona University
- Rachel Umpleby, student at Purdue University, converted to staff

PUBLICATION:

- Cooper, Justin, et al., "Developing A Digital Twin of a TRUEX Extraction Process that Enables Nonproliferation and Safeguards Monitoring through Optical Spectroscopy and Machine Learning," Nuclear Science and Technology Open Research Journal, 2024.

INTELLECTUAL PROPERTY:

- Katherine Jesse, "Deep Lynx MATLAB Adapter," 2023.



The general architecture of the solvent extraction centrifugal cascade digital twin. This includes data collected via ultraviolet-visible (UV/Viz) spectrum sensors which are uploaded to DeepLynx. That data is then made available for artificial intelligence and computational applications. Anomaly detection and concentration predictions and the analytical results are stored in DeepLynx. Separately, the MATLAB adapter runs MATLAB and Simulink ordinary differential equation (ODE) solvers to predict the concentration of metals present in the aqueous and organic solutions at each contactor under steady-state conditions. Finally, information is communicated to the user through visual display via both a web interface and extended reality interface.

Development of a multi-sensor data science system used for signature development on solvent extraction processes conducted for the Beartooth Test Bed

PROJECT NUMBER:
22A1059-085FP

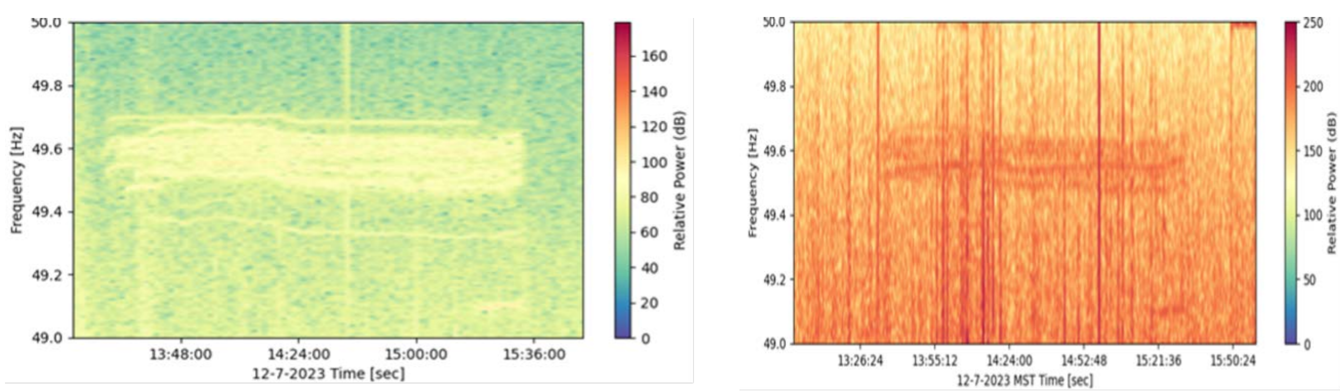
TOTAL APPROVED AMOUNT:
$1,784,419 over 3 years

PRINCIPAL INVESTIGATOR:
Edna Cárdenas

CO-INVESTIGATORS:
Cody Walker, INL
Jay Hix, INL
Katherine Wilsdon Jesse, INL
Luis Ocampo Giraldo, INL
Mitchell Greenhalgh, INL

Novel solvent extraction system monitoring technologies elevate operational performance, provide operators with vital insights, and enable the more precise and comprehensive digital twin creation and safeguards approaches.

Solvent extraction closes the nuclear fuel cycle by enabling the separation and reuse of elements from waste products. Traditional monitoring of a solvent extraction system has been reliant on manual sampling followed by post-process laboratory analysis, a method that is both time-consuming and delays decision-making. This project evaluated the effectiveness of non-traditional sensors in providing informative signals of process or equipment events that would allow for the fine-tuning of operations in a timely manner. Concurrently, machine learning methods were explored to identify anomalies and assess predictive maintenance capabilities. The approach involved the temporary integration of a range of unconventional sensors, ones not typically associated with solvent extraction equipment, and the orchestration of targeted events to gauge sensor capabilities and signal relevance. The correlation of these targeted events with sensor outputs confirmed that non-traditional sensors can capture meaningful signals during solvent extraction operations. Notably, acoustic and seismic sensors measured the operational rate of centrifugal contactors, accelerometers provided insights into pump flow rate ratios, color sensors have the potential to estimate solution concentrations, and infrared cameras paired with advanced machine learning techniques proved effective at detecting leaks. Furthermore, some of the non-traditional sensors examined in this project have been incorporated into the design of INL's solvent extraction testbed, Beartooth. This research has already yielded tangible intellectual property outcomes, including a provisional patent and a decision to assert software copyright for a novel automated leak detection system using infrared cameras.



Acoustic spectrograms showing signals measured 14 meters (left) and 40 meters and outside the facility (right) from contactor one in the cascade. Signals show promise for remote detection of contactor operations.

TALENT PIPELINE:

- Amari Garrett, student at North Carolina Agricultural and Technical State University
- Anne-Marie Boseman, student at Dillard University
- Jay Hix, student at Idaho State University
- Joseph Rivera, student at the University of Puerto Rico
- Kelly Truax, student at University of Hawaii at Manoa
- Melissa Daw, student at College of Eastern Idaho
- Oliver Lewis, student at the University of Liverpool
- Omobolade Odedoyin, student at Johns Hopkins University
- Sarah Popenhagen, student at University of Hawaii at Manoa

PRESENTATIONS AND PUBLICATION:

- E. S. Cárdenas, J. D. Hix, L. A. Ocampo Giraldo, M. R. Greenhalgh, C. M. Walker, and J. T. Johnson, "A new approach to monitoring solvent extraction processes for the nuclear industry," Nuclear Science and Technology Open Research, (2024).
- E. S. Cárdenas, L. A. Ocampo Giraldo, C. M. Walker, M. R. Greenhalgh, J. D. Hix, J. T. Johnson, and K. N. Wilsdon, "A New Approach to Monitoring Solvent Extraction Processes for the Nuclear Industry," 2023 American Nuclear Society Winter Meeting and Technology Expo, Washington, District of Columbia, November 12–15, 2023.
- M. R. Greenhalgh, L. A. Ocampo Giraldo, J. D. Hix, E. S. Cárdenas, C. M. Walker, J. T. Johnson, and K. N. Wilsdon, "Development of a Multi-Sensor Data Science System for Monitoring a Solvent Extraction Process," 46th Actinide Separations Conference, Idaho Falls, Idaho, May 16-17, 2023.
- L. A. Ocampo Giraldo, E. S. Cárdenas, M. A. Garces, M. R. Greenhalgh, J. D. Hix, J. T. Johnson, S. Popenhagen, C. M. Walker, K. N. Wilsdon, "Preliminary Results of a Multi-Sensor Data Science System for Monitoring a Solvent Extraction Process," Institute of Nuclear Materials Management & European Safeguards Research and Development Association 2023 Joint Annual Meeting, Vienna, Austria, May 22-26, 2023.
- C. M. Walker, L. A. Ocampo Giraldo, E. S. Cárdenas, J. D. Hix, J. T. Johnson, M. R. Greenhalgh, and K. N. Wilsdon, "Leak Detection and Sensor Importance within a Solvent Extraction Process." Conference on Data Analysis (2023), Santa Fe, New Mexico, March 7-9, 2023.
- L. A. Ocampo Giraldo, E. S. Cárdenas, J. D. Hix, J. T. Johnson, M. R. Greenhalgh, C. M. Walker and K. N. Wilsdon, "Data Challenges in Multi-Sensor Data Science System for Monitoring a Solvent Extraction Process," Conference on Data Analysis (2023), Santa Fe, New Mexico, March 7-9, 2023.
- E. S. Cárdenas, C. M. Walker, K. Truax, M. R. Greenhalgh, L. A. Ocampo Giraldo, J. D. Hix, J. T. Johnson, and K. N. Wilsdon, "Development of a Multi-Sensor Data Science System for Monitoring a Solvent Extraction Process," Waste Management Symposium (2023), Phoenix, Arizona, February 27-March 3, 2023.
- J. D. Hix, E. S. Cárdenas, L. A. Ocampo Giraldo, "Multi-Sensor Data Acquisition System for Process Monitoring," American Nuclear Society Winter Meeting and Technology Expo, November 13-17, 2022: Phoenix, AZ.
- L. A. Ocampo Giraldo, E. S. Cárdenas, J. D. Hix, M. R. Greenhalgh, C. M. Walker, J. T. Johnson, and K. N. Wilsdon, "Development of a Multi-Sensor Data Science System Used for Signature Development on Solvent Extraction Processes in Support of Safeguards—An Overview," International Atomic Energy Agency Symposium on International Safeguards: Reflecting on the Past and Anticipating the Future, Vienna, Austria, October 31- November 4, 2022.
- C. M. Walker, L. A. Ocampo Giraldo, J. D. Hix, M. R. Greenhalgh, K. N. Wilsdon, E. S. Cárdenas, "Use of Machine Learning for Signature Development in a Multi-Sensor Environment for Safeguard Applications of Solvent Extraction Processes," Sandia National Laboratory 2022 Machine Learning Deep Learning (ML/DL) Workshop, July 25-28, 2022.
- E. S. Cárdenas, L. A. Ocampo Giraldo, J. D. Hix, M. R. Greenhalgh, C. M. Walker, and K. N. Wilsdon, "An Overview in the Development of a Multi-Sensor Data Science System for Monitoring a Solvent Extraction Process," Institute of Nuclear Materials Management 63rd Annual Meeting, July 23-28, 2022.

INTELLECTUAL PROPERTY:

- C. M. Walker, E. S. Cárdenas, L. A. Ocampo Giraldo, J. D. Hix, K. W. Jesse, M. R. Greenhalgh, J. T. Johnson, "A System for Leak Detection and Related Methods," US63/558,444, 2024.

Secondary tracks
Idaho National Laboratory

INTEGRATED ENERGY SYSTEMS

CORE CAPABILITIES

Chemical and Molecular Science

Applied Materials Science and Engineering

Biological and Bioprocess Engineering

Chemical Engineering

Computational Science

Cyber and Information Sciences

Decision Science and Analysis

Earth and Energy Systems and Infrastructure Analysis and Engineering

Nuclear and Radiochemistry

Nuclear Engineering

Systems Engineering and Integration

The lab is pioneering innovative technologies to integrate energy generation, storage, and delivery for a robust energy future. Our multi-scale energy systems research combines nuclear and renewable heat with various forms of electricity generation, including renewables and natural gas, along with carbon capture and sequestration, to accelerate the creation of a secure, reliable and sustainable energy economy while improving grid reliability, resilience and affordability. Our future RD&D efforts will focus on delivering multi-scale integrated energy systems, including nuclear heat and electricity, from basic science to commercial demonstrations. The full life cycle of this research supports the formation of the Energy Technology Proving Ground with megawatt- and megaton-scale test beds. This will allow private sector developers in advanced nuclear, solar, wind, bioenergy, geothermal, and advanced energy carriers, such as hydrogen and ammonia, to test new ideas and technologies to advance the transportation sector, US chemical and manufacturing industries, and the electric grid.

GREGORY HOLMBECK

Innovative carbon dioxide selective membranes from low concentration emission sources

PROJECT NUMBER:
22A1059-070FP

TOTAL APPROVED AMOUNT:
$1,340,000 over 3 years

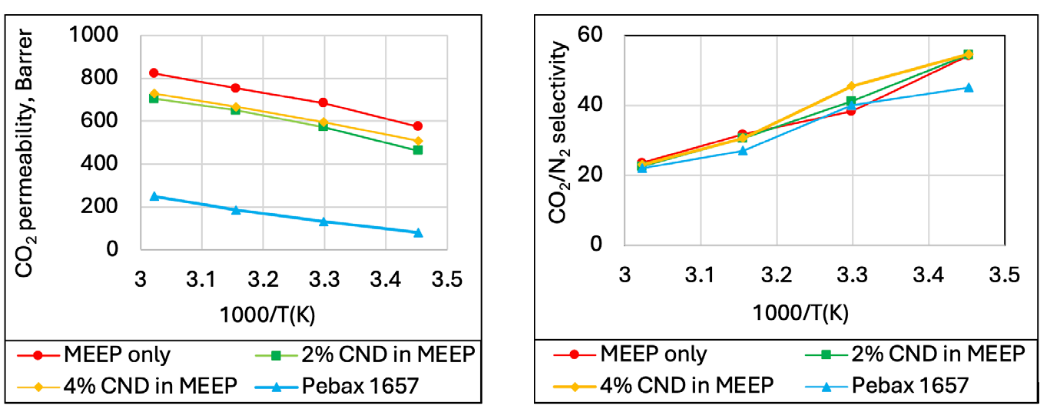
PRINCIPAL INVESTIGATOR:
Birendra Adhikari

CO-INVESTIGATORS:
Christopher Orme, INL
John Klaehn, INL
Joshua McNally, INL

A new cost-effective and environmentally friendly multi-structured membrane advances state-of-the-art carbon capture.

Current state-of-the-art carbon capture technologies, such as cryogenic distillation, solvent-based capture, and pressure swing adsorption, are capital and energy intensive processes. Membrane-based carbon capture is the best alternative because of the low overall cost of capture. However, commercial membranes for these applications often have defects (pinholes) and low carbon dioxide (CO_2) over nitrogen (N_2) selectivity. As a part of this project, INL developed a highly permeable and selective membrane for CO_2 capture from low concentration CO_2 sources that are thin, defect-free, and self-healing. The membrane has a multilayered structure with various support layers, and INL developed a variant of poly[bis(2-(2-methoxyethoxy)ethoxy)phosphazene] (MEEP) as a selective layer. A MEEP selective layer, when filled by 18 carbon atoms (C18) functionalized carbon nanodiamonds (CND), enhances the membrane's mechanical properties. Flat sheets and hollow fiber membranes were developed with different MEEP and CND combinations. INL membranes consistently demonstrated the CO_2/N_2 selectivity of >40 and CO_2 permeability of 450 Barrers at room temperature in all membrane configurations. These separation performances are as good as the state-of-the-art CO_2 selective membranes regarding CO_2/N_2 selectivity and three times better in CO_2 permeability. Techno-economic analysis suggests that these membranes can be used to lower the cost of enrichment and compression to $213 per metric ton of CO_2. Life cycle assessment suggests that these membranes have equivalent CO_2 emissions of 50g of CO_2 emitted per kilogram of CO_2 avoided. These competitive performances suggest that these membranes are applicable in low concentration carbon capture by using low-carbon energy sources, such as nuclear, solar, and wind energy.

INL developed membrane performance compared to commercial Pebax 1657 membrane in terms of (a) CO_2 permeability and (b) CO_2 to N_2 selectivity



(a) (b)

TALENT PIPELINE:

- Amit Nilkar, student at University of Idaho
- Arindam Mukhopadhyay, postdoc at INL, converted to staff
- Hyeonseok Lee, postdoc at INL

PRESENTATIONS AND PUBLICATIONS:

- A. Nilkar, C.J. Orme, J.R. Klaehn, H. Zhao, B. Adhikari, "Life cycle assessment of innovative carbon dioxide selective membranes from low carbon emission sources: a comparative study," Membranes, 13(4), p.410. 2023.
- B. Adhikari, C.J. Orme, J.R. Klaehn, N.S. Nilkar, H. Lee, J.S. McNally, A.D. Wilson, F.F. Stewart, "Cost-effective and environmentally friendly carbon capture from low concentration sources using membranes," 47th Conference on Clean Energy, Clearwater, FL, 2023.
- B. Adhikari, C.J. Orme, J.R. Klaehn, "Techno-economic analysis of carbon dioxide enrichment from low carbon sources using membranes," Chemical Engineering Journal, 2023.
- H. Lee, J.R. Klaehn, C.J. Orme, A.D. Wilson, J.S. McNally, F.F. Stewart, B. Adhikari, "Molecular dynamics study of carbon dioxide, nitrogen, and water permeation through poly[bis((methoxyethoxy)ethoxy)phosphazene] membrane," Chemical Engineering Science, 284, p.119480, 2024.
- B. Adhikari, C.J. Orme, J.R. Klaehn, H. Lee, A.S. Nilkar, H.S. Rollins, A.D. Wilson, F.F. Stewart, "Low-concentration carbon capture using membranes," The North American Membrane Society (NAMS) conference, Tuscaloosa, AL, 2023.
- C.J. Orme, B. Adhikari, J.R. Orme, J.S. McNally, F.F. Stewart, "C18 functionalized nanodiamonds and MEEP-based membranes for efficient CO_2/N2 separation," The NAMS conference, Tuscaloosa, AL, 2023.
- B. Adhikari, C.J. Orme, J.R. Klaehn, J.S. McNally, H. Lee, A.D. Wilson, and F.F. Stewart, "Techno-economic Analysis of Low Concentration Carbon Dioxide Enrichment Using Membranes," Gordon Research Conference, 2022.
- C.J. Orme, B. Adhikari, J.R. Klaehn, J.S. McNally, H. Lee, A.D. Wilson, F.F. Stewart, "Gas Permeability of Carbon Dioxide Selective Phosphazene Polymer Membranes," Gordon Research Conference, 2022.

INTELLECTUAL PROPERTY:

- B. Adhikari, C.J. Orme, J.R. Klaehn, J.S. McNally, H. Lee, F.F.Stewart, "Carbon dioxide selective membranes, gas separation systems including the carbon dioxide selective membranes, and related methods," US patent WO2023220743A2 (2023).

AWARD:

- 2nd place best poster award, 2024 LDRD projects at INL

Optimized microreactor operations with liquid metal battery electrical and thermal energy storage devices

PROJECT NUMBER:
22A1059-086FP

TOTAL APPROVED AMOUNT:
$695,945 over 3 years

PRINCIPAL INVESTIGATOR:
Tyler Westover

CO-INVESTIGATORS:
Amey Shigrekar, INL
Fernando Gallego Dias, INL
Jiangkai Peng, INL
Prabhat Tripathy, INL
Temitayo Olowu, INL
Hojong Kim, Pennsylvania State University

Coupling a liquid metal battery to a microreactor can improve the net present value proposition by 91–141% while improving flexibility, resilience and black start capabilities.

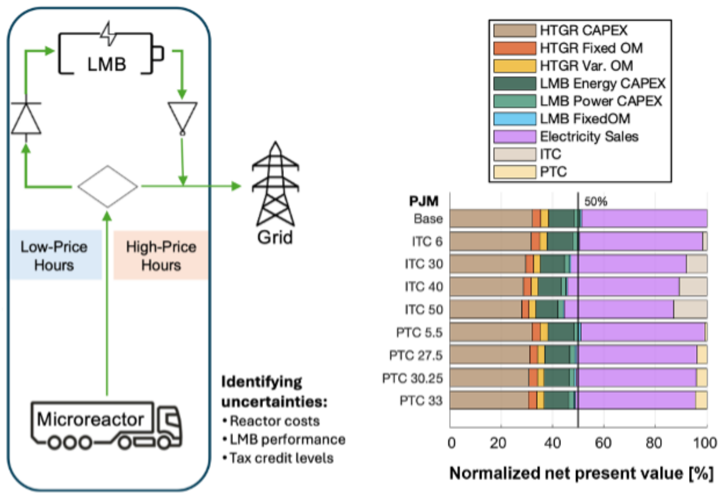
A leading solution to meet carbon emission goals is to tightly couple advanced nuclear reactors to dispatchable industrial thermal and electric power users such that nuclear power can be used for industrial processes when grid electricity demand and prices are low. This project analyzed a system in which a 10 MW microreactor is coupled to a liquid metal battery (LMB) and a high-temperature hydrogen plant in a grid system with high penetration of renewable energy. LMBs can provide extraordinary amounts of current flow, like supercapacitors, because of their liquid material construction and high-temperature operation (450–750°C). These capabilities benefit grid systems with high variable wind and solar power penetration because the fast frequency response and high-power output of the LMB can enhance grid stability. A techno-economic analysis showed that liquid metal batteries can achieve cost parity with lithium-ion batteries due to their construction using low-cost, earth-abundant, and environmentally friendly materials. Laboratory tests were also performed to identify means to further lower the capital cost of LMBs by incorporating lower cost alkali/alkaline earth halide salts with foreign cations such as strontium and potassium. Mixed results were obtained from the laboratory tests indicating that cost saving measures are feasible, but care must be taken to control the electroactive species. The economic performance of an integrated microreactor/LMB system was further analyzed in three markets. Key findings are that coupling an LMB to a microreactor can improve the net present value proposition of a microreactor by as much as 91% without considering potential tax credits and by as much as 141% if tax credits are considered. This work also showed that 24–39% of the microreactor capacity can be utilized for non-electric applications without reducing electricity sales. The wide thermal operating range of LMBs also offers an opportunity for dual use as both electrical and thermal energy storage in the same media. Like other practical batteries, ohmic losses within the cell result in heating. Analyses completed in this project show that heat from the ohmic losses can be captured as thermal energy and stored for later use to further increase the operating efficiency of LMB-coupled systems.

TALENT PIPELINE:

- Jiangkai Peng, postdoc at INL
- Kelly Varnell, student at University of Pennsylvania
- Samuel Root, student at University of Idaho, converted to staff
- So-Bin Cho, student at University of Michigan

PUBLICATIONS:

- K. Varnell, S. Im, P. Asghari-Rad, T. Westover, H. Kim, Assessment of mixed-cation molten salt electrolytes for Li-based liquid metal batteries, Journal of Power Sources, 603, 234387 (2024).
- S. J. Root, A. Shigrekar, T. Westover, Techno-Economic Analysis of a Nuclear Reactor System Coupled with a Liquid Metal Battery, Transactions of the American Nuclear Society, 129(1), 740-743 (2024).
- A. Shigrekar, J. Peng, T. O. Olowu, F. Gallego Dias, T. Westover, Modelling and analysis of nuclear reactor system coupled with a liquid metal battery, The Journal of Engineering (5), e12382 (2024).



System impacts of tax credits on the microreactor-LMB system. The microreactor is fixed at 20 megawatt-electric (MWe) (9.6 MWe). LMB power and energy capacities are optimized for various market, cost and technology scenarios. Investment tax credit (ITC) rates of 6%, 30%, 40%, and 50% are shown on the right, as are production tax credit (PTC) rates of 5.5, 27.5, 30.5, and $33/MWh. Electricity sales indicate net electricity sales after accounting for the costs of grid imports for LMB charging. The black line at 50% net present value distinguishes scenarios that have positive net present value. For example, if the sum of electricity sales and tax credits sales exceeds 50% of the total, the scenario has a net present value greater than 0.

Nuclear-renewable-storage digital twin: enhancing design, dispatch, and cyber response of integrated energy systems

PROJECT NUMBER:
22A1059-112FP

TOTAL APPROVED AMOUNT:
$1,615,000 over 3 years

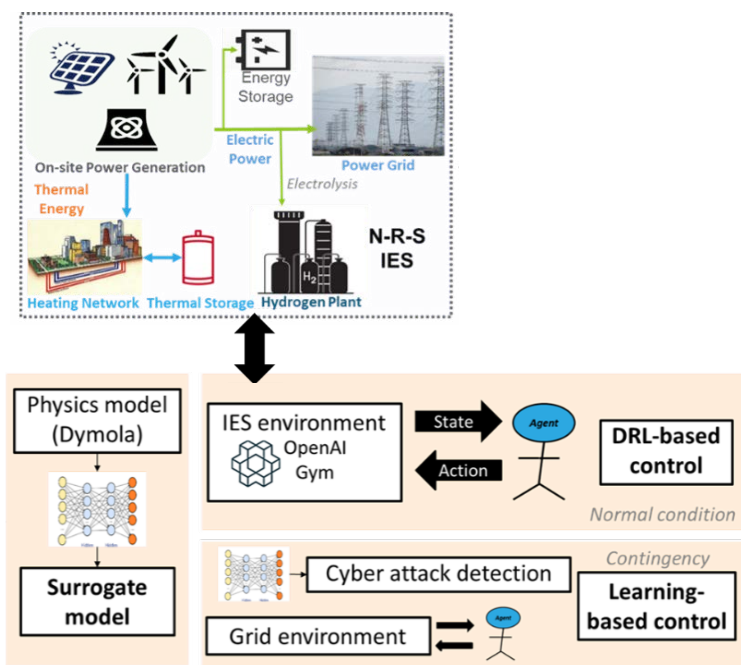
PRINCIPAL INVESTIGATOR:
Binghui Li

CO-INVESTIGATORS:
Bikash Poudel, INL
Christopher Ritter, INL
Jianqiao Huang, INL
Maria Eduarda Montezzo Coelho, INL
Mukesh Gautam, INL
Paul Talbot, INL
Shannon Leigh Eggers, INL
Thomas Mosier, INL
Timothy McJunkin, INL
Jie Zhang, University of Texas at Dallas

COLLABORATOR:
University of Texas at Dallas

Learning-based algorithms making decisions for nuclear-renewable-storage integrated energy systems improve grid economic, reliability, and resilience performance.

This project developed a learning-based and digital twin enabled modeling and simulation framework for economic and resilient real-time decision-making of physics-informed integrated energy system operations. The team first developed high-fidelity physics models of nuclear-renewable-storage integrated energy system by integrating components from several open source Modelica libraries and conducted a 24-hour quasi-static simulation to demonstrate the viability of using integrated energy system to meet end-use heat demands. Based on the physics model, we then developed a set of surrogate models to efficiently represent the quasi-static response of integrated energy system components. The surrogate models were then used in conjunction with deep reinforcement learning algorithms to determine the optimal setpoints of all integrated energy system components for both optimal economic performance and autonomous load following control. To minimize the operation costs of the integrated energy system in an electricity market setting, we formulated a bilevel economic dispatch model to simultaneously optimize its set points and bidding strategy. For risk mitigation under disruptive events, we developed a cyberattack detection technique by using generative artificial intelligence and an intentional islanding strategy by using graph reinforcement learning and demonstrated their viability and efficacy on several testbed systems. Finally, to enable real-time communication across all modules, we developed a data warehouse to store operations data and completed multiple data threads. The results demonstrated advantage of the learning-based decision-making framework for both economic operations under normal conditions and risk mitigation under contingencies.



This project consists of three modules: (1) High-fidelity physics models and surrogate model representations. (2) Optimal control and decision-making based on deep reinforcement learning. (3) Detection and mitigation of disruptive events based on generative artificial intelligence and learning-based control.

TALENT PIPELINE:

- Bikash Poudel, postdoc at INL, converted to staff
- Jiangkai Peng, postdoc at INL
- Jianqiao Huang, postdoc at INL
- Jingbo Wang, student at University of Texas at Dallas
- Mukesh Gautam, postdoc at INL
- Roshni Anna Jacob, student at University of Texas at Dallas
- Sobhan Badakhshan, student at University of Texas at Dallas

PRESENTATIONS AND PUBLICATIONS:

- Poudel, Bikash, et al., "Design, modeling and simulation of nuclear-powered integrated energy systems with cascaded heating applications," Journal of Renewable and Sustainable Energy 15.5 (2023).
- Jacob, Roshni Anna, and Jie Zhang, "Modeling and control of nuclear-renewable integrated energy systems: Dynamic system model for green electricity and hydrogen production," Journal of Renewable and Sustainable Energy 15.4 (2023).
- Rahman, Jubeyer and Jie Zhang, "Steady-state Modeling of Small Modular Reactors for Multi-timescale Power System Operations with Temporally Coupled Sub-models," IEEE Transactions on Power Systems (2024).
- Gautam, Mukesh, Bikash Poudel, and Binghui Li, "Data-Driven Quasi-Static Surrogate Models for Nuclear-Powered Integrated Energy Systems," 2024 IEEE Texas Power and Energy Conference (TPEC). IEEE, 2024.
- Huang, Jianqiao, et al., "Optimal Operations of Nuclear-Based Integrated Energy Systems with Mixed-Integer Programming," 2024 IEEE TPEC. IEEE, 2024.
- Badakhshan, Sobhan, et al., "Reinforcement Learning for Intentional Islanding in Resilient Power Transmission Systems," 2023 IEEE TPEC. IEEE, 2023.
- Li, B. et al., "Use Probabilistic Forecasts in Reliable and Economic Electricity Market Scheduling and Operations," INFORMS 2022 Annual Meeting, Indianapolis, IN, Oct. 16–19, 2022.
- Jacob, R. and Zhang, J., "Distribution Network Operation with Integrated Energy Systems: Modeling and Control Framework," IEEE Power & Energy Society General Meeting, Orlando, FL, July 16–20, 2023.
- Rahman, J. and Zhang, J., "Steady-state Multi-timescale Modeling and Operation of Small Modular Reactors," IEEE Power & Energy Society General Meeting, Orlando, FL, July 16–20, 2023.
- Badakhshan, S. et al., "Enhancing Grid Resilience Through Intentional Islanding by Reinforcement Learning on Graphs," IEEE Power & Energy Society General Meeting, Orlando, FL, July 16–20, 2023.
- Li, B. et al., "Nuclear-Renewable-Storage Systems: Enhancing Planning and Operations of Integrated Energy Systems," INFORMS 2023 Annual Meeting, Phoenix, AZ, Oct. 15–18, 2023.
- Rahman, Jubeyer, Roshni Anna Jacob, and Jie Zhang. "Multi-timescale power system operations for electrolytic hydrogen generation in integrated nuclear-renewable energy systems." Applied Energy 377 (2025): 124346.

Transient electro-kinetic reactor to accelerate development of advanced catalysts for electrocatalytic carbon dioxide reduction to ethylene

PROJECT NUMBER:
23A1070-091FP

TOTAL APPROVED AMOUNT:
$750,000 over 2 years

PRINCIPAL INVESTIGATOR:
Rebecca Fushimi

CO-INVESTIGATOR:
Debtanu Maiti, INL

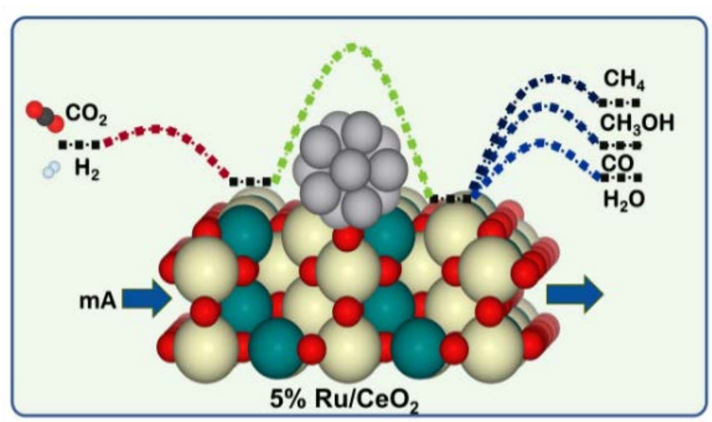
COLLABORATORS:
Washington University in Saint Louis
Webster University

Electric field and dynamic reactant feed modulation enhances carbon dioxide hydrogenation.

Direct electrification approaches for chemical manufacturing decarbonization are a must for net-zero global emissions from chemical industries. CO_2 hydrogenation is an important chemical reaction that enables sustainable conversion of CO_2 to high-value hydrocarbons. Some of the major products obtained through this reaction are methane (CH_4), methanol (CH_3OH), and syngas. Subsequently, this syngas or methanol can be converted to ethylene via Fischer-Tropsch or methanol-to-olefins processes. Application of dynamic catalysis principles in the form of external electric field and hydrogen (H_2) flowrate modulation can pave the way for energy-efficient conversion of waste CO_2 to valuable products at lower temperatures. These processes also enable chemical manufacturing integration with clean hydrogen derived from renewable and sustainable sources. This research demonstrates the application of dynamic electric field and feed modulation for improved reaction performance. CO_2 conversion and methane formation rates were found to increase when the H_2 feed was modulated over a ruthenium impregnated ceria catalyst. Further, the effect of different applied electric waveforms was used to identify enhanced energy efficiency during CO_2 hydrogenation reactions. The synergistic role of dynamic surface concentrations of activated hydrogen and carbon species together with the dynamic charge state of the catalyst surface created a pathway for enhanced CO_2 hydrogenation reaction performance.

PRESENTATION AND PUBLICATION:

- Turaeva, N., Yablonsky, G., Fushimi, R., "Understanding the catalyst 'volcano' dependence through Fermi-level controlled kinetics using electronic theory," Entropy, 2024, submitted, in review.
- Fushimi, R., Turaeva, N., Yablonsky, G., "Fermi-level assisted catalytic kinetics for explanation of volcano-shaped dependence," 18th International Congress on Catalysis, Lyon, France, July 14-19, 2024.



Applying a 1 – 5 mA current to metal catalyst with semiconductor support enhances reactant conversion and governs product selectivity. Modulating the H_2 feed enhances CO_2 conversion and improved CH4 yield.

Advanced instrumentation for in situ analysis and operation of solid oxide electrochemical cells

PROJECT NUMBER:
23P1082-009FP

TOTAL APPROVED AMOUNT:
$149,000 over 1 year

PRINCIPAL INVESTIGATOR:
Nicholas Kane

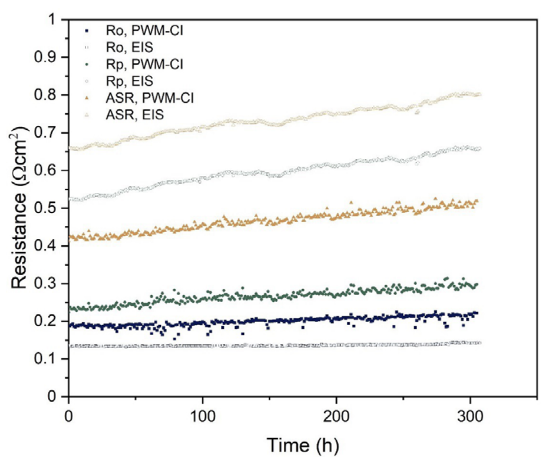
CO-INVESTIGATORS:
Jeremy Hartvigsen, INL
Micah Casteel, INL
Temitayo Olowu, INL

Current interrupt analysis combined with pulse-width modulation control monitors solid oxide electrolysis cell health in real-time.

Solid oxide electrolysis cells will play a critical role in a carbon-free energy future, supplying the green hydrogen feedstock required to decarbonize a myriad of industries. However, several challenges remain to be solved, including the in situ health monitoring of stacks and large area cells. This work developed a new, inexpensive, and fast technique for tracking the health of operating cells. Using pulse-width modulation controls, current interrupt analysis was applied to the cells throughout the test. To perform the experiments, current interrupt control boxes were fabricated, including both low current and high current versions that analyzed button cells and stacks, respectively. A python module analyzed the current interrupts and converted the voltage signals into cell health parameters, including the ohmic, polarization, and area specific resistances. As a demonstration, button cells were tracked over 305 h using both pulse-width modulation–current interrupt analysis and electrochemical impedance spectroscopy. The data show that the pulse-width modulation–current interrupt analysis can effectively track both the ohmic and polarization resistance. Separate analyses using pulse-width modulation–current interrupt and electrochemical impedance spectroscopy demonstrated similar results, showing good agreement between the newly developed technique and the conventional methods. The developed technique expands INL's capability to analyze solid oxide electrolysis cells, providing a deeper analysis into the degradation of the solid oxide electrolysis cells.

TALENT PIPELINE:

- Elvan Sahin, student Virginia Tech
- Nicholas Kane, postdoc at INL, converted to staff



Cell health parameters, including the ohmic, polarization, and area specific resistance (ASR), as a function of time as measured by pulse-width modulation–current interrupt (PWM-CI, closed markers) and electrochemical impedance spectroscopy (EIS, open markers) techniques.

Proteomic insights to interaction between electroactive bacteria and rare earth elements

PROJECT NUMBER:
23A1070-120FP

TOTAL APPROVED AMOUNT:
$898,265 over 2 years

PRINCIPAL INVESTIGATOR:
Asef Redwan

CO-INVESTIGATORS:
Chelsea St. Germain, INL
David Reed, INL
Yoshiko Fujita, INL

Biomolecules enable rare earth element separation and recovery.

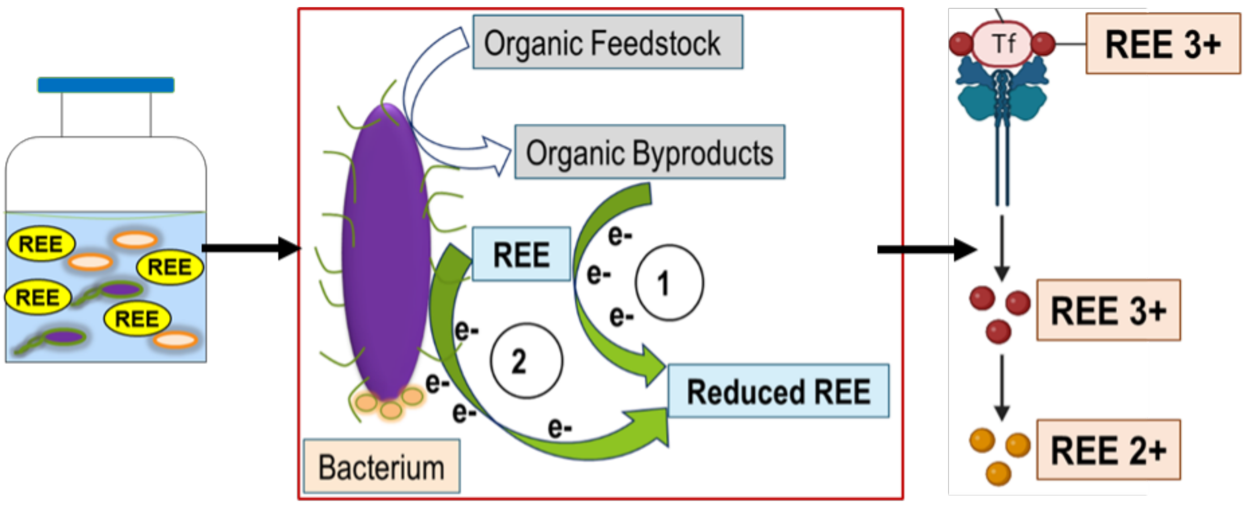
Growth in green energy and other modern technology applications has led to increased interest in supplementing domestic supplies of critically important and valuable metals such as rare earth elements. Because multiple rare earth elements are usually co-recovered from solid matrixes such as ores, electronic wastes, etc., typically of the same oxidation state and similar ionic radius, separations are very challenging. Conventional processes to separate and recover rare earth elements require substantial use of environmentally hazardous chemicals and are energy intensive. New approaches using biological materials or principles have the potential to improve the environmental profile of production. One biological transformation pathway that has been scarcely explored is microbial bioreduction. A member of the *Clostridium* genus of bacteria was reported recently to reduce europium (Eu) from the trivalent to the divalent state, suggesting specific protein interactions with the metal. We postulated that better understanding of the specific biochemical pathways and proteins involved in this process could support the development of novel methods for recovering Eu and likely other rare earth elements from mixed solutions. To elucidate the microbial interactions with Eu at the protein level, we cultured a closely related *Clostridium* strain with different concentrations of Eu(III) and quantified changes in whole organism protein expression levels. We identified metal-binding and electron transfer proteins with differential expression in response to increasing Eu concentrations. Additional analyses revealed that iron-binding, zinc-binding, and cation-binding proteins constituted the largest subset of Eu-responsive proteins. Proteomic fingerprinting identified positive correlation between changes in protein abundances for Eu^{3+} and iron cations in presence of *Clostridium*. *Clostridium* can use amino acids as terminal electron acceptors, but we observed that this process was downregulated when the Eu or iron were added. This observation suggests that Eu(III) or iron(III) can serve as terminal electron acceptor. We also identified upregulation of butyrate metabolism when Eu or iron were present. Furthermore, upregulation of membrane transporters was observed when *Clostridium* was cultured with either 500 µM Eu and iron but not when the bacterium was cultured with the same concentrations of calcium or zinc. These proteins would be key targets for future work to develop bioligands for rare earth element recovery and separation.

TALENT PIPELINE:

- Asef Redwan, postdoc at INL, converted to staff
- Chelsea St. Germain, postdoc at INL
- Jeremy Sabo, postdoc at INL
- Nejar'ye Ivey, student at Bennett College
- Payton Walker, student at Idaho State University

PRESENTATIONS:

- Redwan, A.M., C. St. Germain, G. Morales, M. Walton D.W. Reed, Y. Fujita, "Proteomic insights to interaction between electroactive bacteria and rare earth elements,"The Northwest Regional Meeting (NORM) of the American Chemical Society, Bozeman, Montana, June 2024.
- Redwan, A.M., J.A.A. Sabo, Y. Fujita, D.W. Reed, G. Morales, M. Walton, "Proteomic insights into the interaction between europium and *Clostridium* sporogenes," American Chemical Society Fall Meeting, Denver, Colorado, August 2024.



Conceptual diagram for project, showing bioreduction of the rare earth element (REE) Eu^{3+} by Clostridium and characterization of ion interacting proteins; cartoon on the right shows a protein involved in transport of metal ions into the cell.

Rational design of three-dimensional porous carbon architecture for efficient carbon dioxide capture with high capture capacity and low energy input for regeneration

PROJECT NUMBER:
23A1070-166FP

TOTAL APPROVED AMOUNT:
$650,000 over 2 years

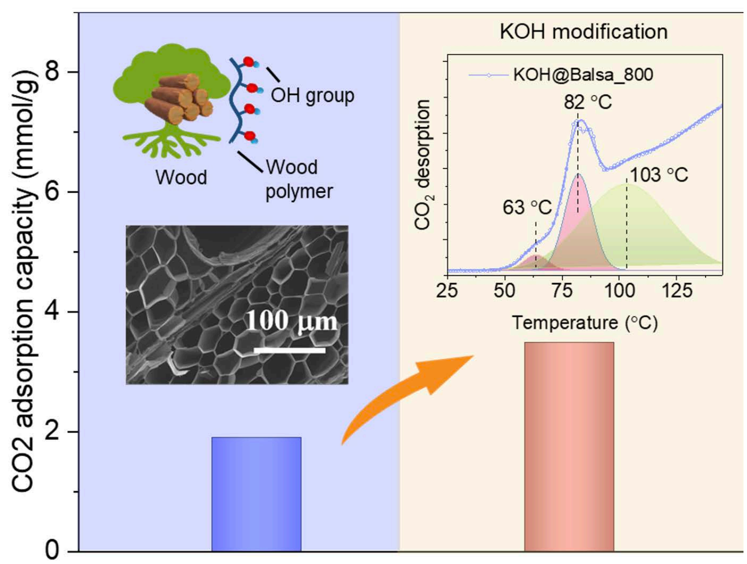
PRINCIPAL INVESTIGATOR:
Yuqing Meng

CO-INVESTIGATORS:
Lucun Wang, INL
Seth Snyder, INL
Haiyan Zhao, University of Idaho
Jin Qian, Lawrence Berkeley National Laboratory
Yingchao Yang, University of Missouri

COLLABORATOR:
University of Maine

Efficient and sustainable carbon capture system developed by combining experimental and theoretical approaches.

Developing highly efficient sorbent systems with both high CO_2 adsorption efficiency and ease of regeneration remains a challenge in direct air capture. In this project, a novel approach for CO_2 capture was developed through designing a three-dimensional porous carbon architecture based on a readily available biomass resource—balsa wood or other carbon materials. Highly porous carbon materials were successfully synthesized by high-temperature pyrolysis followed by surface engineering via introducing abundant surface functional groups as well as growing nanostructures that provide additional adsorption sites for CO_2. The effect of various factors including surface treatment methods, temperature, and CO_2 concentration on CO_2 adsorption capacity and kinetics were systematically investigated. Among various treated balsa carbon materials, the alkali treated sample exhibits the highest CO_2 adsorption capacity with a chemisorption-dominated mechanism as revealed by the infrared spectroscopy characterization. Complementary computational modeling was also performed to accelerate the discovery of novel, highly efficient carbon-based materials for CO_2 capture by calculating the CO_2 adsorption energy on various doped, functionalized, and defect graphene surfaces. Overall, the combined experimental and theoretical approach enables a greater scientific understanding for how to manipulate the sorbent surface chemistry and develop operational conditions to enhance CO_2 capture and regeneration.



Carbon dioxide sorption performance and mechanism of balsa carbon materials.

TALENT PIPELINE:

- Asmita Jana, postdoc at Lawrence Berkeley National Laboratory
- Drew Glenna, student at University of Idaho
- Manish Neupane, student at University of Missouri
- Yuqing Meng, postdoc at INL

PUBLICATIONS:

- A. Jana, S.W. Snyder, E.J. Crumlin, and Jin Qian, "Integrated carbon capture and conversion: A review on C2+ product mechanisms and mechanism-guided strategies," Frontiers in Chemistry 2023, 11, 1-11.
- D.M. Glenna, A. Jana, Q. Xu, Y. Wang, Y. Meng, Y. Yang, M. Neupane, L. Wang, H. Zhao, J. Qian, and S.W. Snyder, "Carbon capture: theoretical guidelines for activated carbon-based CO_2 adsorption material evaluation," Journal of Physical Chemistry Letters 2023, 14, 47, 10693–10699.

INTELLECTUAL PROPERTY:

- Y. Wang, S.W. Snyder, Y. Meng, L. Wang, H. Zhao, D. Glenna, J. Qian, A. Jana, M. Neupane, Y. Yang, "Apparatus, systems, and methods for using 3D porous carbon architecture for efficient carbon dioxides capture with high capture capacity and low energy input for regeneration," Provisional Patent Application No. 63/673,334.

Moderate high pressure x-ray radiation-induced degradation of persistent poly- and perfluoroalkyl substances

PROJECT NUMBER:
23P1079-010FP

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

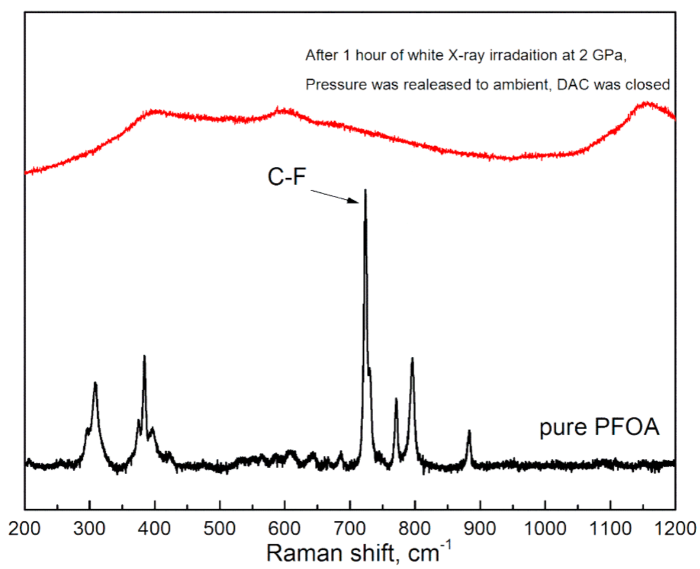
PRINCIPAL INVESTIGATOR:
Gregory Holmbeck

CO-INVESTIGATORS:
Egor Evlyukhin,
University of Nevada Las Vegas
Michael Pravica,
University of Nevada Las Vegas

COLLABORATORS:
Army Research Laboratory
Canadian Light Source

First-of-a-kind high pressure irradiation experiments capture the crystal structure and radiolytic behavior of perfluorononanoic acid—an important perfluoroalkyl substance in environmental contamination.

Poly- and perfluoroalkyl substances (PFAS) are manufactured chemicals that contain at least one carbon-fluorine (C–F) bond. Their extreme chemical resistance has led to PFAS accumulation and persistence in the environment. These compounds have also been linked to several physiological problems, including infertility and cancer. Consequently, a significant research effort has been devoted to the development of efficient PFAS treatment technologies. To date, many of these technologies have not been very successful due to the extreme chemical stability of the C–F bond. Here, we investigated the proof-of-concept of whether moderate high pressure ($\leq$ 10 GPa) can enhance the extent of ionizing radiation-induced PFAS destruction. By using synchrotron x-rays or intense ultraviolet irradiation, we found that perfluorononanoic acid (PFNA) and perflurooctanoic (PFOA) exhibited enhanced degradation when pressurized in a diamond anvil cell (DAC) up to 30 GPa. These first-of-a-kind experiments were also supported by establishing the crystal structure of PFNA, missing information needed to accurately determine the loss of PFNA with irradiation at pressure. These x-ray diffraction data were obtained from beamline 04ID-2 at the Canadian Light Source. Overall, this project accomplished its objective by semi-quantitatively establishing that the combined application of moderate high pressures and ionizing radiation fields promote enhanced PFAS degradation.



Raman spectra (532 nm) of PFOA pressurized to 2 GPa before (black) and after synchrotron x-ray irradiation (red) demonstrating radiation-induced molecular breakdown of PFOA.

TALENT PIPELINE:

- Alejandro Aceves, student at University of Nevada Las Vegas
- Egor Evlyukhin, postdoc at University of Nevada Las Vegas
- Petrika Cifligu, student at University of Nevada Las Vegas
- Trimaan Malik, student at University of Nevada Las Vegas
- William Rogers, student at University of Nevada Las Vegas

PUBLICATION:

- J.W. Reid, T. Malik, M.G. Pravica, A.F.G. Leontowich, and A. Rahemtulla, "Crystal Structure of Perfluorononanoic Acid ($C_9HF_{17}O_2$)," Journal of Powder Diffraction 2024, First View, 1–7.

AWARD:

- Beamtime was secured at: (i) the Lawrence Berkely National Laboratory Advanced Light Source, "Studies of inner shell chemistry under extreme conditions," proposal ALS-12460; and (ii) the CLS, "Studies of per- and poly-fluorinated compounds at extreme conditions," proposal 37G13034~Pravica.

Feasibility study of integrated carbon mineralization and stimulated hydrogen production in Eastern Snake River Plain

PROJECT NUMBER:
23P1080-011FP

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PRINCIPAL INVESTIGATOR:
Wencheng Jin

CO-INVESTIGATORS:
Dong Ding, INL
Ghanashyam Hari Neupane, INL
Robert Egert, INL
Zeyu Zhao, INL

A combination of thermal, chemical, and mechanical stimulation enables geological hydrogen production at the wellhead.

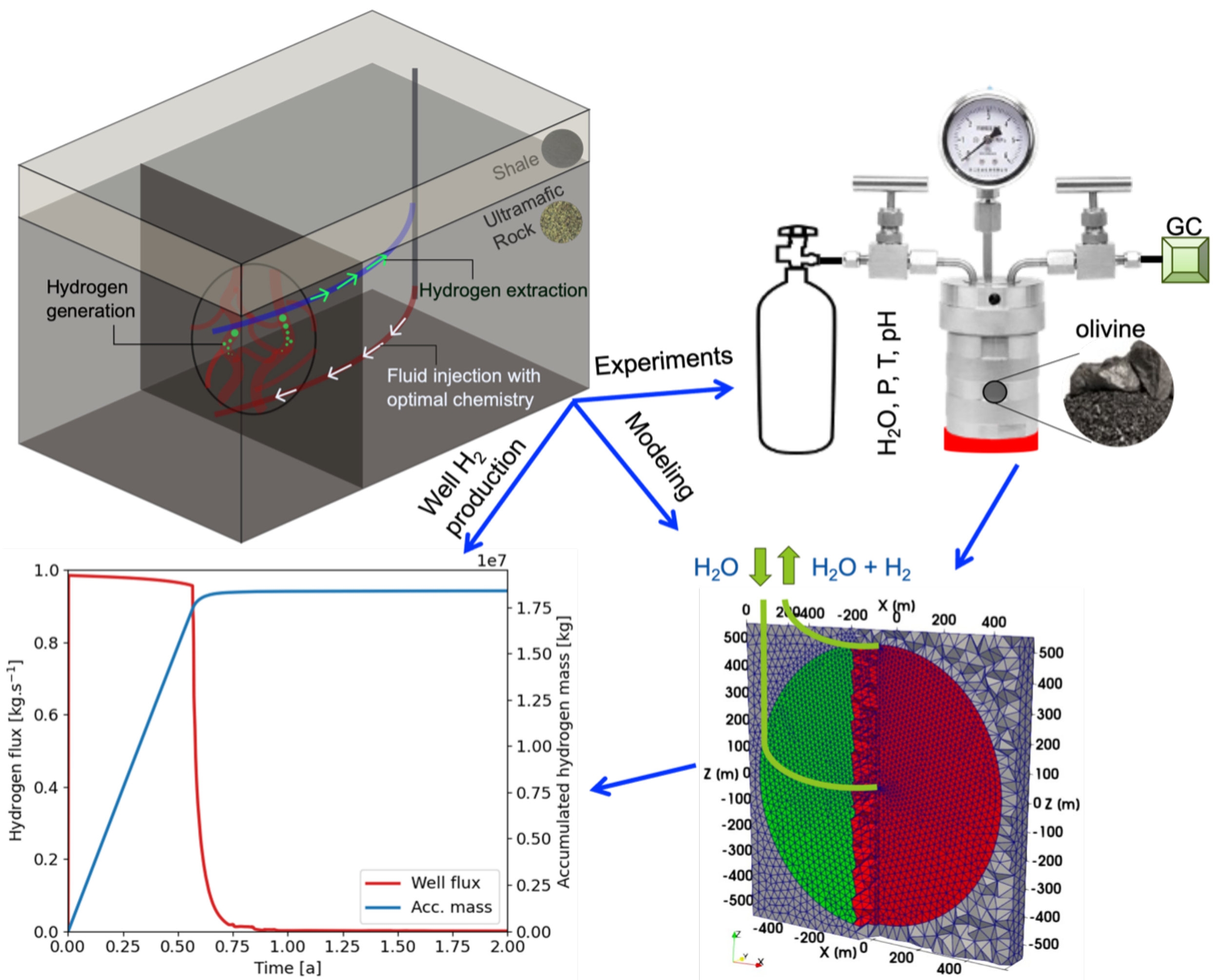
The discovery of geologic hydrogen reserves holding a promise of immense resources for clean energy has ignited a global surge in natural hydrogen exploration. Notably, harvesting clean geologic hydrogen by stimulating hydrogen-producing reactions has a great potential to meet the US DOE's hydrogen Earthshot goal of $1 per kilogram in 1 decade. To unleash this great potential, two fundamental questions need to be addressed: 1) Can a >104 hydrogen generation rate increase be achieved by stimulation? 2) What hydrogen production rate and quantity can be produced at the wellhead? We addressed the first question by experimental characterization of the hydrogen generation rate from olivine serpentinization. 1mm size olivine sands were reacted with water at atmospheric pressure and 95°C in iron-free reactors for 90 days. Both hydrogen sensors and gas chromatography were used to quantify the amount of hydrogen in the header space of the reactor throughout the experimental duration. The results show a 1x10-10 (vol. fraction/sec) hydrogen generate rate, which is several orders of magnitude higher than the field observation from the ophiolite formation in Oman. This high rate resulted from pure olivine reaction at a large surface area and the continuous depleting of hydrogen from the fluid space to the header space. The second question was addressed by numerical simulation of hydrogen migration due to stimulation in saturated formation with varying porosity. Our findings indicate that key parameters, such as stimulation zone permeability and width, significantly impact hydrogen production. High permeabilities combined with large widths can lead to high hydrogen production rates, reaching up to 1 kg/s at the wellhead for rock volume. Additionally, the availability of ferrous iron, rather than the serpentinization rate, is identified as the primary limiting factor for economically viable hydrogen production. While an unstimulated reservoir yields only 45 tons of hydrogen in two years, various stimulation techniques can increase production to 18,500 tons. These results demonstrate that stimulated geological hydrogen production is viable, but significant research and development effort with advanced experiments and modeling is required to realize its full potential.

TALENT PIPELINE:

- Robert Egert, postdoc at INL
- Vuong Van Pham, student at University of Kansas
- Zeyu Zhao, postdoc at INL, converted to staff

PRESENTATIONS:

- Jin, Wencheng, Zeyu Zhao, Robert Egert, Ghanashyam Neupane and Dong Ding, "On the Viability of Stimulated Hydrogen Generation from Iron-rich Formations," AGU Fall Meeting Abstracts, vol. 2024.
- Jin, Wencheng, Dong Ding, Robert Egert, Ghanashyam H. Neupane, and Zeyu Zhao, "Feasibility Study of Integrated Carbon Mineralization and Stimulated Hydrogen Production in Eastern Snake River Plain," AGU Fall Meeting Abstracts, vol. 2023, no. 529, pp. NS13C-0529. 2023.



An integrated approach combining experiments and numerical simulations was established to predict geological hydrogen production from ultramafic formation. The results show a significant amount of hydrogen production at the wellhead with novel stimulation technologies.


CAUTION
INL
Idaho National Laboratory

ADVANCED MATERIALS AND MANUFACTURING FOR EXTREME ENVIRONMENTS

CORE CAPABILITIES

Applied Mathematics

Applied Materials Science & Engineering

Computational Science

Condensed Matter Physics and Materials Science

Earth and Energy Systems and Infrastructure Analysis and Engineering

Mechanical Design and Engineering

Nuclear Engineering

Advanced materials with improved performance in extreme environments are critical to achieve our clean energy and security objectives. Therefore, the lab is actively working to shift the paradigm from design-build-test to digital design and manufacturing for nuclear fuels, lightweight materials and advanced survivability materials. To achieve this, we are leveraging existing expertise in metallurgy, materials and materials performance in extreme environments with innovation in advanced manufacturing and AI. Our work will enable next generation materials performance using AI coupled with advanced manufacturing to predict materials degradation ranging from the microscale to the mesoscale in a wide range of extreme service environments with process-informed physics.

MICHAEL MOOREHEAD

Construction of a unified discontinuous Galerkin finite element framework for simulation of fracture network evolution in multiphysics systems

PROJECT NUMBER:
21P1058-001FP

TOTAL APPROVED AMOUNT:
$827,911 over 3 years

PRINCIPAL INVESTIGATOR:
Wencheng Jin

CO-INVESTIGATOR:
Ruijie Liu, INL

Newly developed software simulates hydraulic fracturing with subsurface proppant transport.

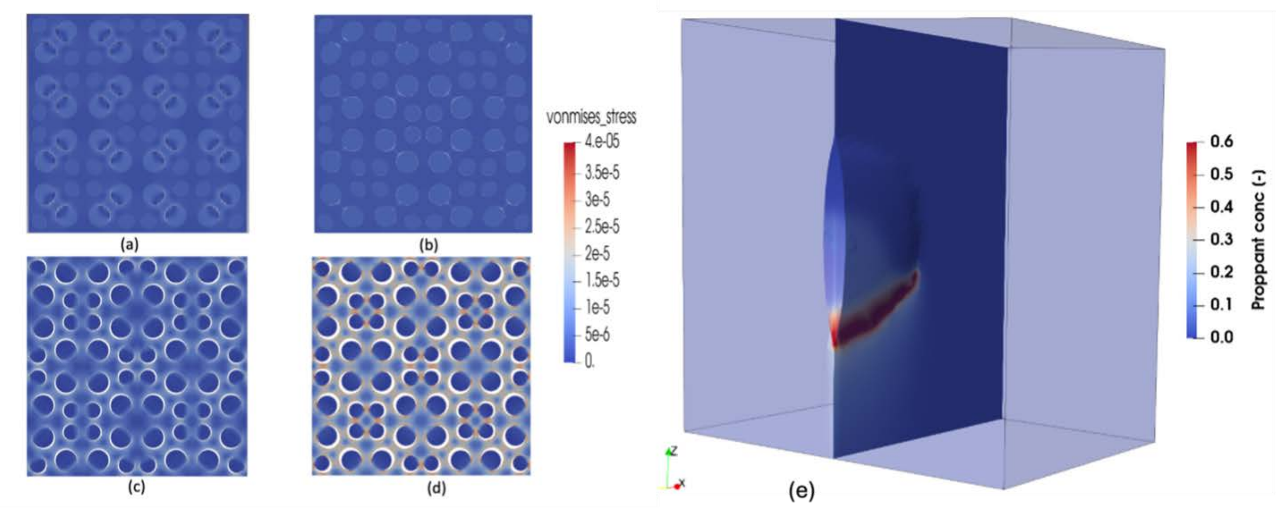
Hydraulic fracturing in porous media challenges computational frameworks in robustly and efficiently handling difficult fracture propagation, abrupt fluid flow within narrow fractures, abrupt and highly localized fluid pressure loading on fracture aperture wall surfaces, and coupling and interaction of the solid and fluid phase fields. Fracture propagation results in new paired internal surface boundaries and fluid flow within narrow fractures has been validated by using the lubricating theory formulated on the corresponding lower dimensional and intermediate interface in the middle of the paired aperture surfaces. Regarding these surface-oriented physical events, we employed discontinuous Galerkin finite element framework for the formulations on governing equations. Specifically, to improve the instability potentially occurring at the stage of crack nucleation, we modified the existing Incomplete Interior Penalty Galerkin scheme to generate a gap rather than a penetration across discontinuous Galerkin interfaces when interfaces are under tension. Furthermore, we transferred the formulation for fluid flow on the intermediate interface of a fracture aperture to its corresponding paired and existing fracture wall solid element surfaces. By such a transformation, we avoided additional mesh generation for these intermediate interfaces sitting in the middle of the narrow fracture apertures. Following the verification of our formulation using Khristianovich-Geertsma-DeKlerk benchmarks, we constructed a five-crack model to further demonstrate the capability and performance of our proposed computational framework by studying the interaction between human-made and natural fractures in a fractured reservoir subject to fluid injection. We further extended the hydraulic fracturing modeling capabilities by incorporating proppant transport and settling within the propagating fracture. These capabilities enable the application of subsurface stimulation in enhanced geothermal systems, oil and gas industry, and in situ leaching of critical minerals.

TALENT PIPELINE:

- Robert Egert, postdoc at INL

PRESENTATIONS AND PUBLICATIONS:

- Liu, R., Jin, W., Harbour, L. H., Kong, F., Permann, C. J., Gaston, D. R., & Podgorney, R. K., "An IIPG-based finite element framework in MOOSE for modeling fiber reinforced composite failure governed by extrinsic cohesive laws," International Conference on Physics of Reactors 2022 (PHYSOR 2022), Pittsburgh, PA, May 15–20, 2022.
- Liu, R., Jin, W., Harbour, L., Kong, F., Permann, C., Gaston, D., & Podgorney, R. (2023). A robust interface finite element formulation for modeling brittle material failure problems. International Journal for Numerical Methods in Engineering, 124(23), 5356-5374.
- Egert, R. W., Jin, W., Fournier, A., & Meng, C., "A Novel Workflow for Coupled Simulation of Hydraulic Stimulation with Simultaneous Injection of Proppant, Proceedings, 49th Workshop on Geothermal Reservoir Engineering Stanford University, Stanford, California, February 12–14, 2024.



Kitfox predicted nucleation, propagation, and arrest of cracks, following bilateral displacement loading increase for fiber-reinforcement composites model with 64 fibers at (a) u = 0.545 m, (b) u = 0.565 m, (c) u = 0.96 m, and (d) u = 2.0 m. (e) Predicted proppant transport and settlement in three-dimensional propagating fracture.

Advanced material property prediction through digital twins

PROJECT NUMBER:
22A1059-044FP

TOTAL APPROVED AMOUNT:
$1,590,000 over 3 years

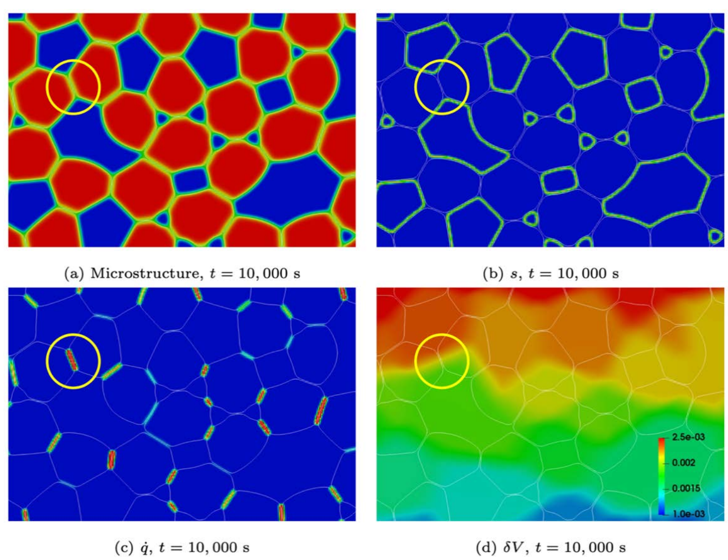
PRINCIPAL INVESTIGATOR:
Brennan Harris

CO-INVESTIGATORS:
Donna Post Guillen, INL
Juan Gallego Calderon, INL
Larry Aagesen, INL
Rajiv Khadka, INL
Stephanie Pitts, INL
Lucas Robinson, Purdue University
R. Edwin Garcia, Purdue University

COLLABORATORS:
Boise State University
Massachusetts Institute of Technology

A new digital framework to organize manufacturing data and generate experiment designs shortens the design life cycle and automates process-property response surface model creation.

Traditional manufacturing methods such as forging, casting, and milling have been in practice for hundreds or thousands of years. Modern practitioners of these methods have an abundance of accumulated knowledge to optimize their products. Manufacturing methods that have emerged or reached maturity more recently, such as electric field-assisted sintering and digital light processing, lack the same deep well of literature and experience to use for tuning their process parameters for optimal design. This project developed a modern digital approach to informing the selection of advanced manufacturing parameters for enhanced sample performance, targeted at accelerating the maturity of these new methods. The team produced a granular digital representation of manufacturing processes, parameters, geometry, and materials. This manufacturing ontology, called Advanced Manufacturing Basis Entity Relationships, is open source and reusable in other digital twin and simulation efforts in the manufacturing space. The project also created an application that uses Advanced Manufacturing Basis Entity Relationships as a backend for gathering manufacturing data that resembles a lab notebook interface. The application, called Advanced Methods for Manufacturing using Ontology and Numeric Objects for Iterative Design, created predictive response surface models for alumina and yttria-stabilized zirconia samples manufactured via digital light processing. The process has also been used to define a digital twin of the electric field-assisted sintering method for copper, tungsten, boron carbide, and yttria.



(a) Microstructure of the zoomed-in region the center of the simulation domain at $t = 10,000$ s. (b) Electrical conductivity s, (c) local heat generation rate $\dot{q}$, and (d) applied field δV of the zoomed-in region at $t = 10,000$ s. The contour of $\lambda = 0.65$ is overlaid on each image to show the corresponding particle microstructure. The color bar used for (d) differs from the others to allow finer resolution of details of the potential drop. The heat generation is predominantly localized to grain boundaries, as illustrated by the yellow-circled grain boundary in each image.

TALENT PIPELINE:

- Danny Hermawan, student at Purdue University
- Jonathan Sampson, student at Massachusetts Institute of Technology
- Lucas Robinson, student at Purdue University
- Soumya Sarangi, student at Purdue University
- Timothy Bragg, student at Boise State University

PUBLICATIONS:

- Aagesen, L., Pitts, S., & Garcia, E. (n.d.), "Electrochemical grand potential-based phase-field simulation of electric field assisted sintering," Acta Materialia, 120049, August 2024.
- Harris, B., Post Guillen, D., Monson, A., "Digital Light Processing of Yttria-stabilized Zirconia: Modeling Photoinitiator Decay," Frontiers in Energy Research, vol. 12, September 2024.

INTELLECTUAL PROPERTY:

- Harris, B. Advanced Manufacturing Basic Entity Relationships (AMBER) [software ontology], August 2024, https://github.com/idaholab/AMBER

Tailoring the properties of multiphase materials through the use of correlative microscopy and machine learning

PROJECT NUMBER:
22A1059-068FP

TOTAL APPROVED AMOUNT:
$1,000,000 over 2 years

PRINCIPAL INVESTIGATOR:
Thomas Mason

CO-INVESTIGATORS:
Colin Merriman, INL
Jason Walleser, INL
K. Ryan Bratton, INL
David Field, Washington State University

COLLABORATORS:
Carnegie Mellon University
Pacific Northwest National Laboratory

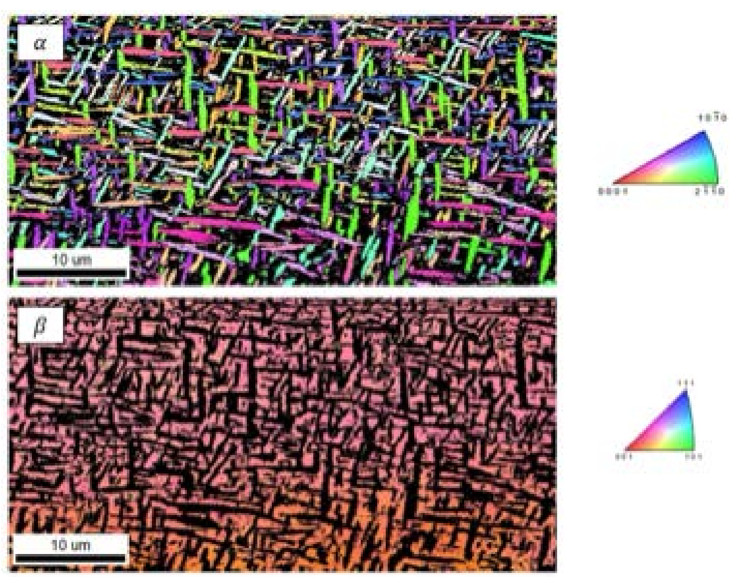
A deep learning neural network segmented and classified the phases of complex multiphase materials in support of further materials optimization efforts.

The goal of this project was to implement an automated analysis capability that could use multiple disparate streams of material characterization data to generate a better understanding of the roles that microstructural details play in macroscopic performance. In particular, the team sought to use varied measurements from optical microscopy, electron backscatter diffraction, energy dispersive spectroscopy and scanning electron microscopy to provide multilayer, quantitative ground truth measures of the microstructures that could then be fed through common, state-of-the-art, machine learning routines to identify local details about dominant gain shapes or neighbor relationships to add insight to failure characterizations. This project aimed to identify and correlate the critical microstructural features in a titanium-10vanadium-2iron-3aluminum alloy that is reported to simultaneously exhibit high strength and fracture toughness. These data were used to train a neural network in a semi-supervised environment to identify key microstructural features such as platelet dimensions and locations and phase boundaries and correlate those features with the strength and toughness.

This work included machine learning tool development and material thermomechanical processing and characterization efforts. The team imported a machine learning tool from the literature, modified it to meet local requirements, and successfully implemented it through supervised training. This tool is currently under use in the laboratory today. The material processing and characterization was less successful as the selected alloy retains a high density of dislocations (imperfections) in its lattice after the initial processing steps. This high dislocation density adversely affects the preferred characterization technique using electron diffraction backscatter resulting in low quality grain maps for subsequent analysis.

TALENT PIPELINE:

- Gunnar Blaschke, student at Washington State University



Reconstructed grain maps from electron backscatter diffraction measurements. The two maps are highlighted according to the two phases present—α and β—with their corresponding inverse pole figure providing information about the orientation of each pixel in the map. Each image was taken at 4000× magnification.

High-throughput spark plasma sintering of compositional arrays

PROJECT NUMBER:
22P1066-007FP

TOTAL APPROVED AMOUNT:
$346,800 over 3 years

PRINCIPAL INVESTIGATOR:
Michael Moorehead

CO-INVESTIGATORS:
Alex Pomo, INL
Allen Roach, INL
Arin Preston, INL
Jorgen Rufner, INL

Sample array featuring 19 unique high entropy alloy compositions produced simultaneously using the newly developed parallelized electric field assisted sintering process.

New high-throughput electric field assisted sintering manufacturing technique produced over 100 unique high entropy alloy compositions in a fraction of the time needed by traditional methodologies.

Many industrial applications taking place in harsh service environments are limited by the performance of their materials. However, the process of discovering and developing new materials can be expensive and time-consuming. To accelerate this process, INL researchers developed a new high-throughput synthesis technique, enabling many samples of varying compositions and geometries to be produced simultaneously using electric field assisted sintering. Traditional electric field assisted sintering involves placing a single powder charge into an electrically conductive die and punch set. Electric current is passed between the punches, which heats the material as the punches are compressed, densifying the powder into a single part. The new process employs the use of custom electric field assisted sintering tooling, which features an array of recesses that can each be filled with a different powder charge. A volume of pressure transfer media is added above the custom tooling, which maintains uniform pressure on each of the recesses while sintering, even as the powder charges densify at different times during the process. The result is an array of fully dense samples that are either attached to a substrate or left isolated depending on the choice of pressure transfer media. This parallelized electric field assisted sintering technique was used to produce over 100 unique high entropy alloy compositions in substantially less time than traditional manufacturing techniques. The arrays of high entropy alloy compositions were polished and characterized in parallel using scanning electron microscopy and energy dispersive spectroscopy. The high entropy alloy arrays were subsequently sent to the University of Wisconsin–Madison Ion Beam Laboratory where they were subjected to heavy ion irradiation up to 50 displacements per atom (dpa) at 600°C to screen for promising alloy compositions for advanced nuclear applications.

TALENT PIPELINE:

- Arin Preston, postdoc at INL, converted to staff
- Michael Moorehead, Russell L. Heath distinguished postdoctoral fellow at INL, converted to staff

PRESENTATION:

- M. Moorehead, A. Preston, Z. Hua, J. Rufner, High-Throughput Electric Field Assisted Sintering and Characterization Techniques for Materials Discovery, TMS, San Diego, CA, 2023.

INTELLECTUAL PROPERTY:

- Michael Moorehead, Jorgen Rufner, Arin Preston, "Methods of Forming Sintered Articles and Associated Assemblies and Components," application no. PCT/US23/73565

Development of an in situ instrument for real-time microstructure monitoring of advanced manufacturing processes

PROJECT NUMBER:
22A1059-142FP

TOTAL APPROVED AMOUNT:
$750,000 over 3 years

PRINCIPAL INVESTIGATOR:
Zilong Hua

CO-INVESTIGATORS:
Amey Khanolkar, INL
Asa Monson, INL
Caleb Picklesimer, INL
Jorgen Rufner, INL
Michael McMurtrey, INL
Robert Schley, INL

Lock-in thermography identifies transport property and microstructure defects of advanced manufacturing produced parts in operando.

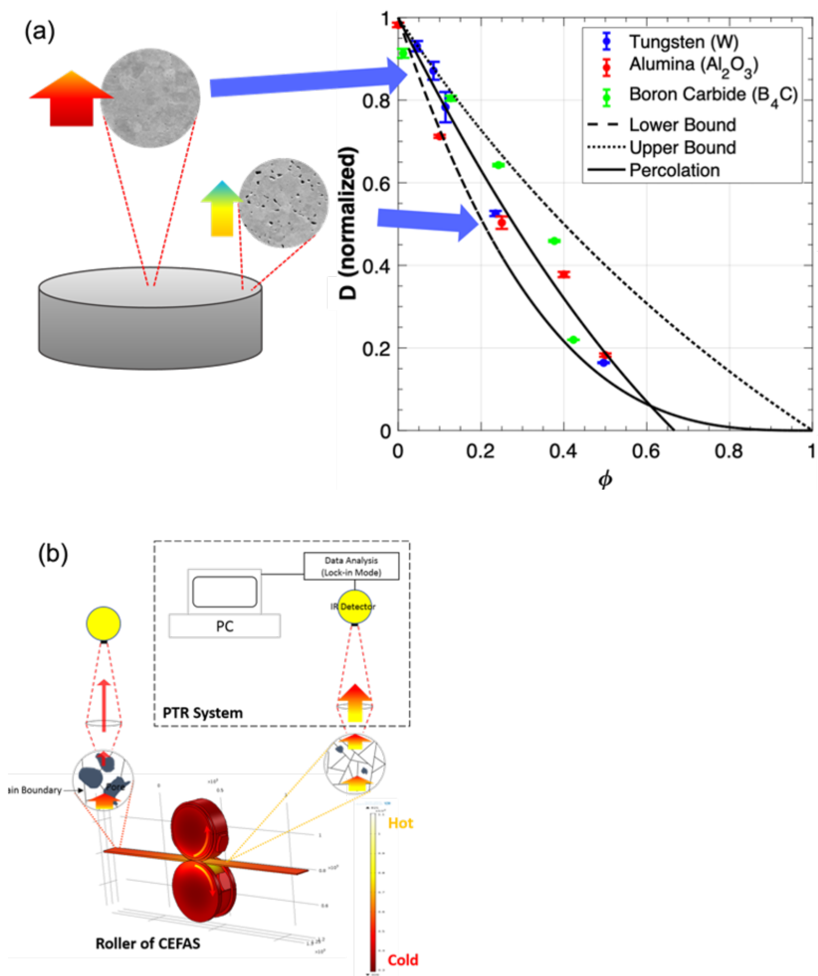
Advanced Manufacturing technology can produce materials with complex geometry and versatility, enabling the development of novel functional materials in a cost-effective and efficient way. However, rapid microstructure characterization of advanced manufacturing parts during the manufacturing process or in operando remains a technical gap. The discovery, fabrication and quality assurance of novel materials is still a trial-and-error procedure. In this project, the team validated the correlation between thermal diffusivity and microstructure defects in advanced manufacturing manufactured materials with the focus on the local porosity, then developed a lock-in thermography based system that can rapidly screen the thermal diffusivity of the advanced manufacturing parts. As a laser-based technique, lock-in thermography measurement is remote and nondestructive, and it is thus feasible to deploy the measurement system to limited spaces. Thermal wave-based measurements are capable of handling thermal anisotropy, which is necessary for investigating the anisotropic pores/keyholes that is generally observed in advanced manufacturing parts. Meanwhile, as it collects blackbody radiation as the probe, lock-in thermography works particularly well on materials with industrial-grade rough surfaces and at elevated temperatures. Most importantly, by using a million-pixel lock-in camera to replace the traditional infrared detector, we can equivalently conduct several traditional thermal wave measurements in parallel, greatly reducing the experiment time from hours to seconds. Using lock-in thermography, we measured a series of direct energy deposition manufactured materials with various manufacturing conditions and identified that the direct energy deposition laser power is most impactful to the formation of porosity, followed by the powder feed rate and laser scan velocity. This lock-in thermography system is ready to be deployed to INL's unique Continuous Electric Field Assisted Sintering with the module design completed and module built.

TALENT PIPELINE:

- Isabella Stepanek, student at Georgia Institute of Technology
- Patrick Merighe, student at Utah State University, INL internship helped get into the PhD program at Georgia Institute of Technology

PUBLICATIONS:

- Z. Hua, P. Merighe, J. Rufner, A. Preston, R. Schley, Y. Wang, S. Doran, and D. Hurley, "Microstructure characterization of electric field assisted sintering (EFAS) sintered metallic and ceramic materials using local thermal diffusivity measurement," AIP Advances, 13, 095220 (2023).
- A. Negi, H. P. Kim, Z. Hua, A. Timofeeva, X. Zhang, Y. Zhu, K. Peters, D. Kumah, X. Jiang, and J. Liu, "Ferroelectric Domain Wall Engineering Enables Thermal Modulation in PMN–PT Single Crystals," Advanced Materials, 35, 22, 2211286 (2023).



(a) Thermal diffusivity is closely correlated to the local density/porosity. Consequently, the microstructure defect information can be revealed by measuring thermal diffusivity. (b) The design diagram of deploying the thermal diffusivity measurement system to Continuous Electric Field Assisted Sintering.

Complex internal structures in castings for extreme environment applications

PROJECT NUMBER:
23A1070-146FP

TOTAL APPROVED AMOUNT:
$636,300 over 2 years

PRINCIPAL INVESTIGATOR:
Michael Mulholland

CO-INVESTIGATORS:
Thomas Lillo, INL
Zherui Guo, INL

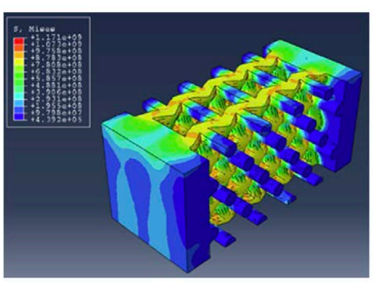
COLLABORATOR:
Brigham Young University

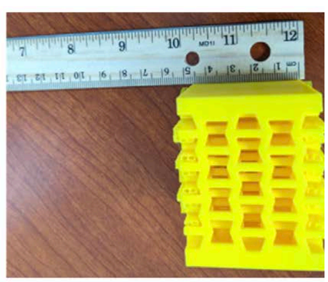
Investment casting coupled with fused deposition method printing enables fabrication of complex auxetic steel lattice structures for lightweight armor applications.

Developing lightweight vehicles is important in both civilian and military applications to improve fuel efficiency and extend range. One way to decrease the weight of military vehicles is to decrease the weight of the protective armor system while maintaining the same level of protection. Lattice structures can reduce weight and maintain strength and performance. Auxetic lattice structures offer superior energy absorption due to their ability to densify while being compressed. Complex cast lattice structures were fabricated using additive manufacturing to make the molds at a higher throughput as compared to laser powder bed fusion. Ongoing research is conducting the first experimental ballistic test of complex auxetic structures. The project successfully produced cast steel complex auxetic lattice structures using investment casting coupled with fused deposition method printing of the casting mold patterns. Finite element modeling was used to predict the quasi-static compressive response and ballistic response of the lattice structures and to optimize the lattice design. Two different steels were cast and a third is in progress. The project also modeled the castability of the three different alloys, and the microstructure was examined to qualitatively verify the model predictions. Experimental ballistic testing and quasi-static compression testing are still in progress. The project produced heat exchangers using the same method, but the size and design of heat exchangers prohibited the use of the investment casting approach.

TALENT PIPELINE:

- Jonathon Handy, student at Brigham Young University
- Joshua Lim, student at Brigham Young University


(a)


(b)


(c)

(a) Abaqus model of quasi-static compression of the lattice structure; (b) plastic fused deposition method printed lattice pattern; and (c) cast auxetic 316L steel lattice structure.

Cost-effective transparent lithium aluminum oxynitride ceramics

PROJECT NUMBER:
23P1075-017FP

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

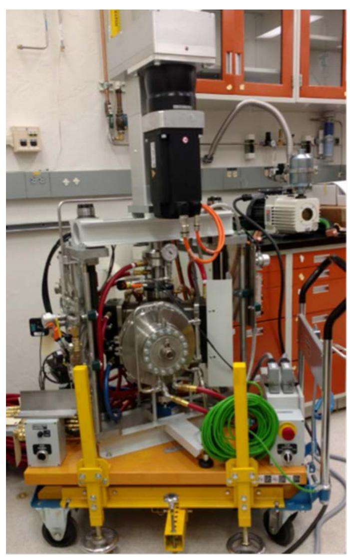
PRINCIPAL INVESTIGATOR:
Michael Mulholland

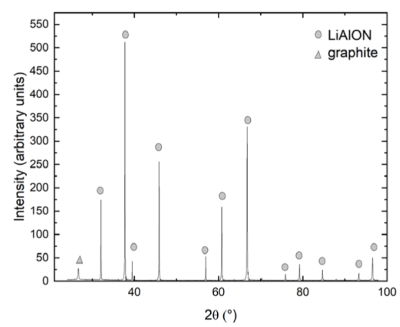
CO-INVESTIGATORS:
Jorgen Rufner, INL
Michael Glazoff, INL

Reactive electric field assisted sintering enables efficient synthesis of lithium aluminum oxynitride ceramics without pre-reacted powder.

Transparent ceramics are highly sought-after for applications ranging from lighting to lasers to high temperature high erosion resistance windows for harsh environments. The last application is of considerable interest for windshields on aircraft, missiles, and vehicles. The wide use of transparent aluminum oxynitride in window applications is limited by its high cost and long fabrication times. This project reduced the cost of aluminum oxynitride by simplifying the process and by reducing the process time using electric field assisted sintering with a lithium aluminate sintering aid. The project developed the parameters to reactively sinter a ball-milled mixture of aluminum oxide, aluminum nitride, and lithium aluminate into phase-pure cubic lithium aluminum oxynitride (LiAlON) without pre-reacted powder. The result was achieved after only 5 minutes of sintering at 1800°C, a more than two orders of magnitude improvement as compared to the conventional process time of ~20 hours at >1850°C. Transparency was not yet achieved due to an interaction between residual porosity and carbon contamination from the graphite tooling. The LiALON was chosen partially due to the hypothesis that it would reduce carbon contamination, but it did not reduce contamination enough to prevent the LiALON samples from turning black.

Reactive electric field assisted sintering using the Thermal Technologies direct current sintering unit (a) produces phase-pure LiAlON as shown by the x-ray diffraction pattern in (b).





Synthesis of cubic gallium nitride templates and heterostructures

PROJECT NUMBER:
23P1075-011FP

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

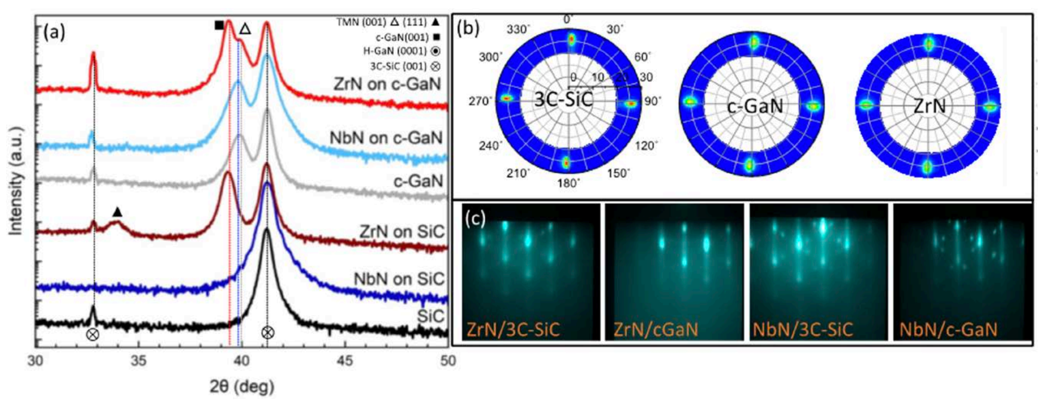
PRINCIPAL INVESTIGATOR:
Brelon May

CO-INVESTIGATOR:
Kevin Vallejo, INL

COLLABORATORS:
Montana State University
The Ohio State University

New epitaxial growth of metastable cubic gallium nitride and superconducting transition metal nitride thin films enables applications in quantum science and optical devices.

Researchers developed single crystalline (100) cubic gallium nitride (c-GaN) growth on 3C silicon carbide (SiC) templates and subsequent epitaxial films of zirconium nitride (ZrN) and niobium nitride (NbN) by molecular beam epitaxy. It was found that an in situ substrate cleaning procedure and shutter sequencing yielded purely the metastable c-GaN phase and completely suppressed formation of the stable hexagonal polytype, even at elevated temperatures. Detailed x-ray diffraction and transmission electron microscopy measurements confirm the epitaxial nature of both the c-GaN and transition metal nitride layers. Transmission electron microscopy also reveals that the three-dimensional growth of the c-GaN layer results in large area roughness. However, the transition metal nitrides conformally cover this layer, and the interface is extremely sharp. The transition metal nitride thin films also show superconducting transitions at <10 K, lower than their bulk counterparts. Compared to growth directly on 3C-SiC and hexagonal GaN templates, the growths on c-GaN layers showed worse transport properties. The lower critical temperatures are likely due to increased defects, roughness, and impurities, which could all be improved with further study. This work provides a platform for the epitaxial integration of a cubic wide bandgap semiconductor with superconducting metals, which could have a variety of potential applications from quantum science to optical devices.



(a) X-ray diffraction 2θ-ω scans of the template, c-GaN on template, and metal nitrides directly on the template and on c-GaN. (b) X-ray diffraction pole figures of the {113} peak of the template, c-GaN, and ZrN verify the azimuthal relationship between substrate and film. (c) reflection high energy electron diffraction images taken at the end of growth for the superconducting films.

TALENT PIPELINE:

- John (Pierce) Fix, student at Montana State University
- Kevin Vallejo, Russell L. Heath distinguished postdoctoral fellow at INL, converted to staff
- Nicole Fessler, student at Cornell University
- Trent Garrett, student at Boise State University
- Zach Cresswell, student at University of Minnesota

PRESENTATIONS AND PUBLICATION:

- Zach Cresswell, Sabin Regmi, Anshul Kamboj, Kaustubh Bawane, Boopathy Kombaiah, Kevin Vallejo, Nicole Fessler, Tent Garrett, Paul Simmonds, Krzysztof Gofryk, Brelon J. May, "Epitaxial integration of cubic GaN and superconducting nitrides," Nano Letters.
- Brelon J. May, "Molecular Beam epitaxy of Superconducting Zirconium nitride thin films on GaN substrates," American Vacuum Society (AVS), November 2023.
- Zach Cresswell, "Epitaxial growth of cubic GaN and ZrN," American Physical Society (APS), March 2024.
- Brelon J. May, "Molecular beam epitaxy of cubic nitrides," Invited presentation at TMS, March 2024.
- Zach Cresswell, "Epitaxial integration of transition metal nitrides with cubic gallium nitride," North American MBE (NAMBE), June 2024.
- Brelon J. May, "Epitaxial synthesis and integration of cubic nitrides," Invited presentation at Pacific Northwest (PNW) AVS, August 2024.

Alloy development for joining materials designed for extreme environments

PROJECT NUMBER:
23P1075-027FP

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

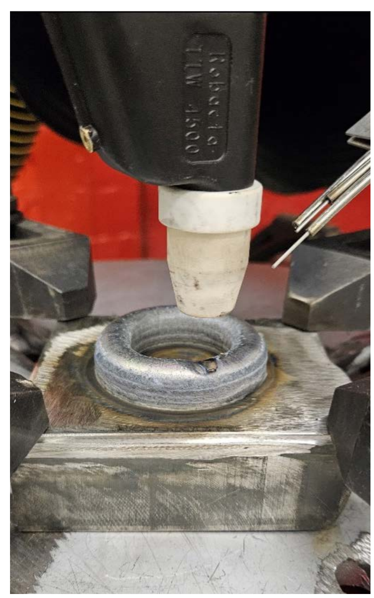
PRINCIPAL INVESTIGATOR:
Tate Patterson

CO-INVESTIGATORS:
Michael Glazoff, INL
Michael Mulholland, INL
Sandeep Dhakal, Boise State University

Solidification temperature range does not proportionally change in relation to the fractions of introduced filler metals.

Prior research has shown that multi-wire arc additive manufacturing can compositionally grade alloys through simultaneous additions of different filler metals. The heat source used in this work was a gas tungsten arc welding power supply/torch with three independently controlled cold-wire feed systems. By using knowledge from Computer Coupling of Phase Diagrams and Thermochemistry (CALPHAD) simulations and the ability to introduce multiple alloys into a weld pool with the multi-wire arc additive manufacturing system, this project showed the ability to rapidly generate new alloy compositions with minimal effort compared to traditional alloy development. This research stemmed from the difficulty of welding Alloy 740H without causing solidification or liquation cracking. The three filler metals used in this investigation included Alloy 740H, Alloy 617, and Alloy 718. These filler metals were chosen because they are nickel-based alloys, are commercially available, and have a useful solidification temperature range. The thermodynamic computations showed that varying the percentages of Alloy 740H, Alloy 617, and Alloy 718 can alter the weld metal composition to reduce crack susceptibility. Although not a sole contributor to crack susceptibility, changes to the solidification temperature range can help determine how susceptible an alloy is to solidification cracking. Computational simulations showed that the solidification temperature range does not proportionally change in relation to the fractions of introduced filler metals. Although the solidification temperature range was lower for Alloy 617 and Alloy 718 compared to Alloy 740H, mixtures of Alloy 740H with 17% Alloy 617 and 17% Alloy 718 maintained the same solidification temperature range. Therefore, it is possible that the crack susceptibility remained the same as the Alloy 740H composition.

Multi-wire arc additive system creating a cylinder by simultaneously introducing multiple wires into the leading edge of the weld pool.



Dynamic scaling analysis of accelerated irradiation testing on additive manufacturing materials with the aid of positron annihilation spectroscopy

PROJECT NUMBER:
23P1080-022FP

TOTAL APPROVED AMOUNT:
$164,700 over 1 year

PRINCIPAL INVESTIGATOR:
Jagoda Urban-Klaehn

CO-INVESTIGATOR:
Tianyi Chen, Oregon State University

Thermally treating three-dimensional printed samples mitigates radiation damage from heavy ion irradiation.

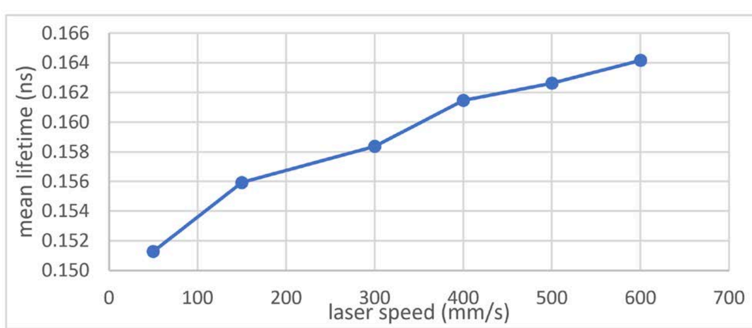
Positron annihilation lifetime spectroscopy was applied to three-dimensional printed stainless steel alloys. These specimens were subsequently irradiated by heavy ions with different doses and conditions to mimic neutron behavior without needing the rigorous safety measures required for neutron irradiation/activation conditions. These experiments were performed to understand the aging process and radiation degradation, especially since these alloys are planned to be used as structural materials for next generation nuclear reactors. Specimens with varied heavy ion irradiation doses after different treatment—as manufactured and thermally treated—were examined. Later, the effects of alloys manufactured with different laser speeds were studied on the nanometer scale in bulk samples. The positron annihilation lifetime spectroscopy studies were complemented by optical spectroscopy and scanning electron microscopy to visualize the surface structure. The positron annihilation lifetime spectroscopy results proved that non-conditioned specimens have much more radiation damage as compared to thermally treated samples. The damage for non-conditioned specimens increased with dose. Manufacturing with high laser speed produced more porosity in the material due to insufficient curing time in the bulk and on the surface. This research proved that positron annihilation lifetime spectroscopy can be used for examination of heavy ion irradiated materials that have lower penetration through the bulk compared to positron as a sensor. They showed that positron annihilation lifetime spectroscopy is a useful spectroscopy for alloys exposed to heavy ion dose, under different thermal pre-treatment or produced with different laser speeds.

TALENT PIPELINE:

- Ian Ferguson, student at Oregon State University
- Johnathan Gonyaw, student at Oregon State University
- Spencer Doran, student at Oregon State University
- Tor Maldonado, student at Oregon State University

PRESENTATION:

- J. Urban-Klaehn, T. Chen, J. K. Conrad, A. Cunningham. Anne Gaffney and all, "Investigation of Gamma/Heavy Ion Irradiated Materials and Nuclear Graphite Studies by use of Positron Annihilation," ANS Transactions, Volume 130, #1, pp. 620-623, June 2024.



Mean lifetime value increases linearly with the laser speed. It shows that positron lifetime spectroscopy is sensitive to the alteration in the microstructure of three-dimensional materials manufactured by different laser speeds, since conditioning matters.

Thermodynamics guided design of refractory structural alloys for extreme environments

PROJECT NUMBER:
23P1082-016FP

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PRINCIPAL INVESTIGATOR:
Sriswaroop Dasari

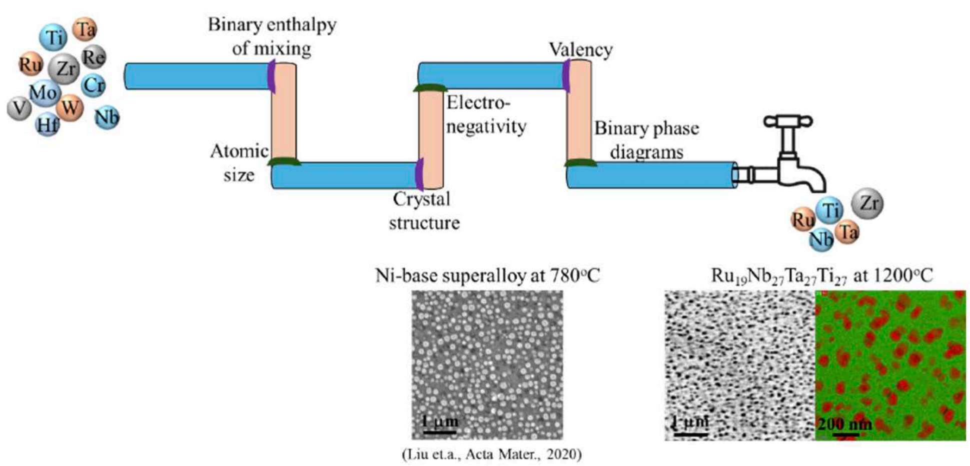
CO-INVESTIGATOR:
Boopathy Kombaiah, INL

COLLABORATOR:
Texas A&M University

Novel refractory alloys can operate at Generation IV nuclear reactor temperatures greater than 800°C.

The next generation nuclear reactors demand structural materials that can withstand extreme irradiation, mechanical, and corrosion/oxidation environments. The materials being used in current generation reactors suffer from degradation phenomena such as high void swelling, poor creep strength, poor oxidation resistance, and embrittlement under such extreme environments. To overcome these limitations, this work developed novel refractory alloys that can meet the goals of advanced nuclear concept reactors. The alloys currently used for extreme temperatures and mechanical loading consist of a mixture of softer solid solution matrix and stronger precipitates. However, obtaining such a microstructure is a challenge in refractory alloys. In this project, we overcame this challenge by employing readily available thermodynamic and first-principles data to design alloys and their microstructures. We hypothesized that ruthenium addition to refractory metals produces a stable mixture of soft body-centered cubic matrix with strong ordered intermetallic precipitates (B2 crystal structure). A binary alloy with 80 atomic percentage (at.%) zirconium and 20 at.% ruthenium and another alloy with 27 at.% each of niobium, tantalum, titanium, and 19 at.% ruthenium were fabricated using arc melting technique in this study. Electron microscopy results showed that both alloys are largely composed of body-centered cubic and B2 phases. The results of this study show the importance of thermodynamics in alloy design for high-temperature applications.

TALENT PIPELINE:
Sriswaroop Dasari, postdoc at INL



Alloy design based on thermodynamic principles leads to the desired precipitation strengthened microstructure for operation beyond 1000°C. Also shown for comparison is a microstructure from one of the nickel-base superalloys, the current 'high-temperature materials' that can be used only up to 1000°C.

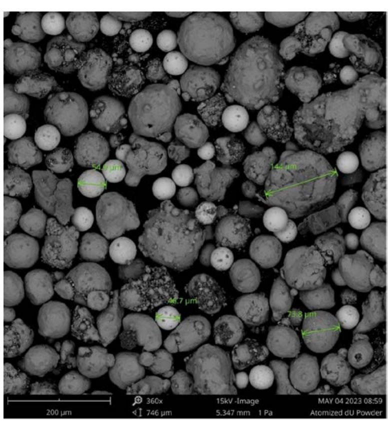
Ultrasonic atomization capability for directed-energy deposition additive manufacturing

PROJECT NUMBER:
23P1082-020FP

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PRINCIPAL INVESTIGATOR:
Joshua Zelina

CO-INVESTIGATOR:
Jennifer Watkins, INL



Depleted uranium powder atomized with ultrasonic atomization/directed-energy deposition method.

Novel ultrasonic atomization capability enables uranium-bearing material fabrication from powder feedstock.

Metal additive manufacturing offers a pathway to fabricating designs that are otherwise un-manufacturable and has the potential to revolutionize many industries, including nuclear. However, the quality of parts fabricated through metal additive manufacturing is dependent on the metal powder feedstock used in the additive manufacturing process. Therefore, fabricating high quality powder feedstock is a fundamental precursor to fabricating high quality parts. For metal additive manufacturing and other advanced manufacturing techniques, atomization is the current state-of-the-art process for producing powder feedstock. Current atomization methods have prohibitively high up-front costs and require specialized facility setups, making it difficult to implement despite fundamentally requiring only three simple components. These components include an energy source to melt bulk feedstock, a secondary energy source to break up the melt into fine droplets, and an inert working atmosphere to prevent melt contamination. Directed-energy deposition additive manufacturing machines, a common metal additive manufacturing method, already have two of these three requirements. This project sought to drastically reduce the up-front cost and installation issues of atomizers by providing the third missing component, a secondary energy source to break apart the melt. This was successfully accomplished by fabricating an ultrasonic atomization component which can be retrofitted into existing directed-energy deposition additive manufacturing machines, thus allowing atomization of bulk feedstock. The ultrasonic atomization component can then be removed to return the directed-energy deposition additive manufacturing machine to its original state. This advanced the state-of-the-art in atomization by reducing the overall cost of an atomizer from hundreds of thousands of dollars or even millions of dollars to ~$20k. Furthermore, installation time was reduced from months for current atomizers to ~30 minutes. Importantly, the ultrasonic atomization/directed-energy deposition approach requires no modifications to the facility or to the directed-energy deposition additive manufacturing machine and takes up no additional footprint.

The ultrasonic atomization/directed-energy deposition setup successfully atomized stainless steel and depleted uranium. The resultant powder is suitable for use in a range of advanced manufacturing methods. This novel capability will support fabrication of uranium powder to meet a wide range of needs. Finally, talks are ongoing with industry partners to determine how best to license and commercialize the intellectual property resulting from this project.

INTELLECTUAL PROPERTY:

- Zelina, J., "Atomization devices for an additive manufacturing apparatus, additive manufacturing systems including an atomization device and methods of atomizing a target substrate," US Patent No. US20240139818A1, 2023.

AWARD:

- 3rd place best poster award, 2024 LDRD projects at INL

SECURE AND RESILIENT CYBER-PHYSICAL SYSTEMS

The lab is securing the nation's critical infrastructure against complex and dynamic cyberthreats by increasing resilience to the full spectrum of hazards and environmental changes. With the rapid expansion of electric vehicle charging systems and power generation from solar and wind resources, there are more unprotected connections to the bulk electric system each year. Moreover, US government officials are increasingly concerned that Chinese state-sponsored cyber actors are seeking to position themselves for disruptive or destructive cyberattacks against US critical infrastructure in the event of a major crisis or conflict. INL already has responded by developing new test beds and simulators for critical infrastructure resiliency R&D and a new range for testing 5G and Next Generation mission critical communications. Over the next five to ten years, we will assist industries, government and academia to adopt the philosophy of Cyber-Informed Engineering and innovate new means for civilian and government entities that provide critical functions to secure themselves against cyberattacks.

ROBERT IVANS

CORE CAPABILITIES

Applied Mathematics

Advanced Computer Science, Visualization, and Data

Cyber and Information Sciences

Decision Science and Analysis

Mechanical Design and Engineering

Nuclear Engineering

Systems Engineering and Integration

Interdependent infrastructure systems resilience analysis for enhanced microreactor power grid penetration

PROJECT NUMBER:
22A1059-069FP

TOTAL APPROVED AMOUNT:
$1,520,000 over 3 years

PRINCIPAL INVESTIGATOR:
Ryan Hruska

CO-INVESTIGATORS:
Bjorn Vaagensmith, INL
Som Dhulipala, INL
Timothy McJunkin, INL
Audrey Olivier, University of Southern California
Sam Yang, Florida State University

Physics-informed graph neural networks methods enhance resilience modeling for utility-scale electric grid planning.

The North American electric grid structure and the interdependent systems that support its operations are evolving and becoming increasingly interconnected. As utilities transition away from centralized large-scale controllable fossil fuel generators to a fleet of smaller non-emitting distributed energy resources, traditional methods used to plan future reliable and resilient electricity delivery are insufficient. Policy makers, regulators and utilities require tools that leverage computationally efficient approaches to evaluate the complex interdependent behaviors within these integrated systems and to understand their potential vulnerabilities to all threats and hazards.

This project developed advanced machine learning methods required to enhance the utility-scale integrated resource planning and grid planning process. The team researched and developed machine learning techniques for power flow and system recovery modeling. The power flow modeling efforts focused on the embedding of uncertainty quantification via Bayesian neural networks and development of physics-informed graph neural networks to integrate dynamic grid topology information. This approach overcomes limitations of traditional and other machine learning based approaches reported on in literature. For example, the generalized graph neural network trained to perform power grid contingency analysis is scalable across multiple grid topologies. This generalized graph neural network model performed better than the direct current power flow approximation when compared to the traditional Newton-Raphson solutions. Furthermore, the generalized graph neural network model only requires a fraction of time to predict the solutions as compared to the Newton-Raphson method.

The team also developed resilience metrics to better assess potential integrated resource planning portfolios beyond the least-cost option. These include an adaptive capacity and expected critical load curtailment metrics. Finally, a prototype hybrid modeling framework was developed to facilitate information exchange between domain load flow models and the discrete event function-failure-logic models.

TALENT PIPELINE:

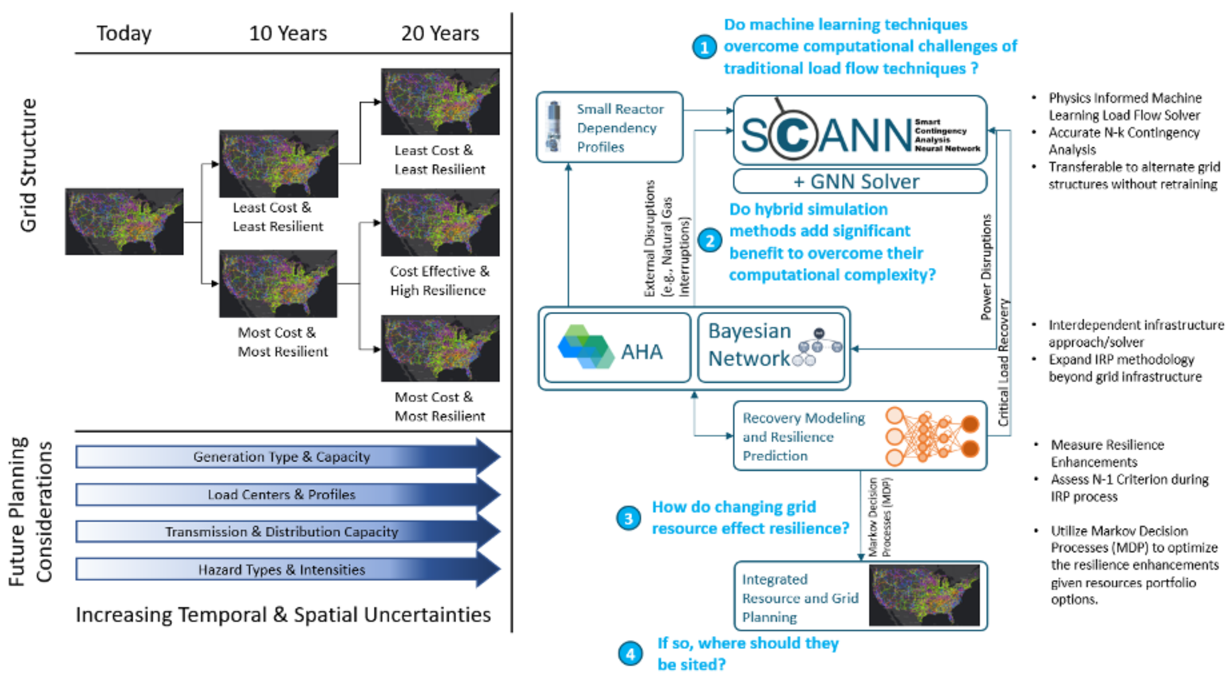
- Nicholas Casaprima, student at University of Southern California
- Mukesh Gautam, postdoc at INL
- Racheal Seidel, student at University of Texas, Austin
- Ryan Lee, student at University of California, Merced
- Tyler Phillips, postdoc at INL, converted to staff

PRESENTATIONS AND PUBLICATION:

- M. Gautam, T. McJunkin, T. Phillips and R. Hruska, "A Resilient Integrated Resource Planning Framework for Transmission Systems: Analysis and Optimization," Sustainability 16.6 (2024): 2449.
- M. Gautam, T. McJunkin, and R. Hruska, "A Resilient Integrated Resource Planning Framework for Transmission Systems: Analysis Using High Impact Low Probability Events," 2024 IEEE Power & Energy Society Innovative Smart Grid Technologies Conference (ISGT). IEEE, 2024.
- S. Yang, B. Vaagensmith, D. Patra, R. Hruska and T. Phillips, "Multi-fidelity power flow solver," 2022 Resilience Week (RWS). IEEE, 2022.
- S. Dhulipala and R. Hruska, "Efficient interdependent systems recovery modeling with DeepONets," 2022 RWS. IEEE, 2022.

INTELLECTUAL PROPERTY:

- B. Vaagensmith, et al., "Smart Contingency Analysis Neural Network (SCANN)" Software Copywrite, INL Technology # CW-23-16, Tracking No. 13006.



Physics-Informed graph neural network-based methodology for enhancing utility-scale integrated resource and grid planning.

Secure information transmission in multi-carrier and multi-antenna fifth generation and beyond wireless communications systems

PROJECT NUMBER:
22A1059-103FP

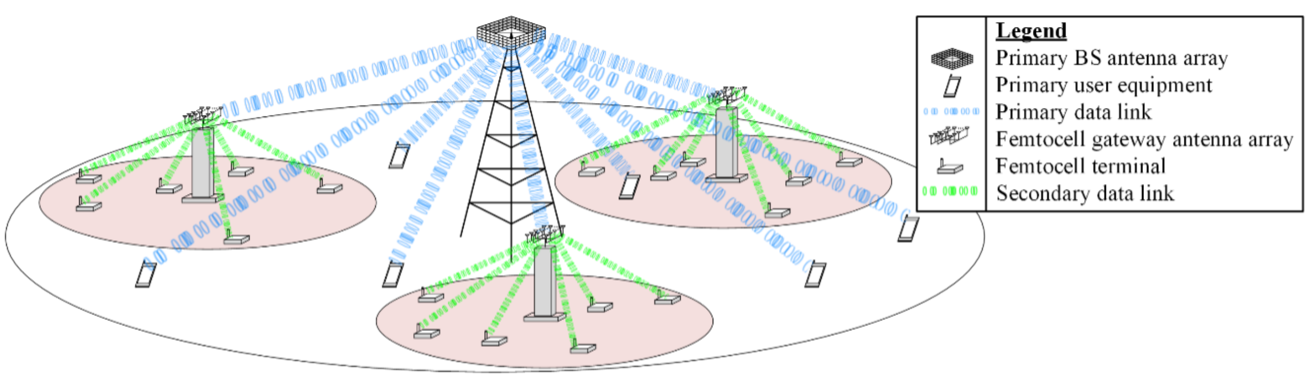
TOTAL APPROVED AMOUNT:
$908,678 over 3 years

PRINCIPAL INVESTIGATOR:
Arslan Majid

CO-INVESTIGATORS:
George Chung Him Yuen, INL
Hussein Moradi, INL
Behrouz Farhang-Boroujeny, University of Utah

Novel signal processing techniques increase 5G and beyond cellular network capacity by at least 80% with a secured end-to-end over-the-air wireless communication.

The research team developed a physical layer security for wireless communication that is recognized as an effective means to enhance wireless security. Our effort was made possible by exploiting the reciprocal nature of the time, frequency, and space varying nature of the wireless channel medium. These properties of wireless channels allowed us to establish a secure communication channel between a pair of users to effectively share a secret—the secret being a realization of the channel—that is statistically uncorrelated for a third party located more than a few wavelengths away from the two users. Related to this research, we took advantage of two prominent lines of research from the prior-art literatures: (i) physical layer key generation, and (ii) secure information transmission. In physical layer key generation, the wireless channel randomness is used to obtain a secret key to be shared between a pair of legitimate parties. The research we pursued relates to the development of secure information transmission techniques. Secure information transmission methods, also referred to as physical layer security, make use of signal processing techniques to degrade the eavesdropper's (Eve) reception quality, while maintaining the link between Alice and Bob (the legitimate parties) above a certain quality. Prior work exists on a multi-carrier based method of introducing artificial noise in a subspace orthogonal to the channel between Alice and Bob, hence blocking Eve from any observation of the communicated information between Alice and Bob. Our research has built off this idea applicable to a broad class of spread-spectrum systems. We also made use of the degrees of freedom provided by multi-antennas to further enhance the secrecy of our designs. The research has opened a new line of research in physical layer security that has not been explored yet. Adding physical layer security to spread-spectrum systems is related to applications which require low probability of detection and are resilient to harsh jamming environments.



Implementation of a cyclic prefixed direct sequence spread-spectrum based femtocell network that synchronizes with 5G+ primary networks and establishes a secondary cyclic prefixed direct sequence spread-spectrum communication network.

TALENT PIPELINE:

- Aaron Pettit, student at University of Utah

PUBLICATIONS:

- B. A. Kenney, A. J. Majid, H. Moradi and B. Farhang-Boroujeny, "Frequency Domain Detection and Precoding for Massive MIMO with Single Carrier Modulation," IEEE Transactions on Wireless Communications, 2022, vol. 21, no. 5, pp. 3232-3248.
- B. Farhang-Boroujeny, A. S. Pettit, A. Majid, H. Moradi, "Physical Layer Security through Cyclic Prefix Direct Sequence Spread Spectrum Signaling," 2022 IEEE Global Communications, May 22, 2022.
- A. S. Pettit, B. Farhang-Boroujeny, H. Moradi, "Security Analysis of a Class of Artificial-Noise-Aided Secured Spread Spectrum Systems," Pending publication in IEEE Transactions on Wireless Communications.

INTELLECTUAL PROPERTY:

- Hussein Moradi, Behrouz Farhang, A. J. Majid, "Spead Spectrum Communication in Femtocell Network, and Associated Devices, Systems, and Methods," Pub No.:US2021/0314020 A1, Oct. 7, 2021.

Adaptive fingerprinting of control system devices through generative adversarial networks

PROJECT NUMBER:
22A1059-116FP

TOTAL APPROVED AMOUNT:
$660,000 over 3 years

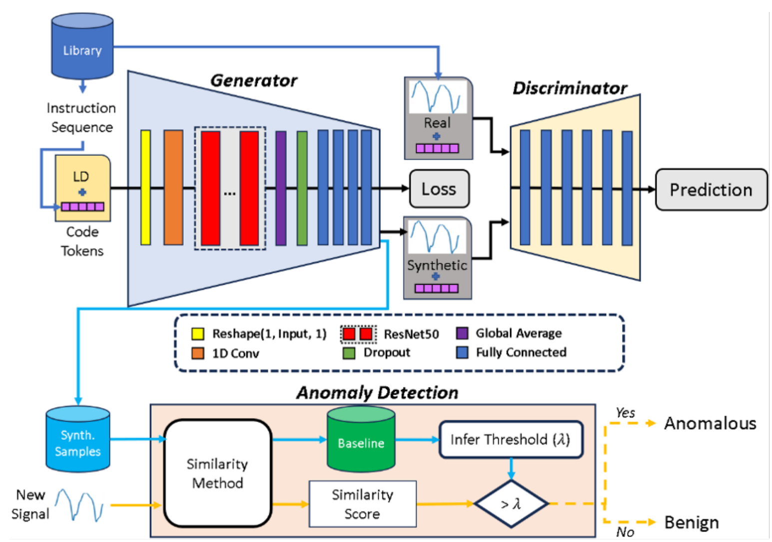
PRINCIPAL INVESTIGATOR:
Robert Ivans

CO-INVESTIGATORS:
Craig Rieger, INL
Constantinos Kolias, University of Idaho

COLLABORATOR:
University of Idaho

Machine learning pipeline elucidates a computer chip's past behavior, predicts its future behavior, and detects anomalous behavior without interfering with its operation.

Having the capability to predict what electromagnetic signals a chip produces for a given known set of operations means having the ability to explain behavior that has happened and the ability to detect unexpected behavior as it happens. This project trained generative adversarial networks to accurately predict involuntarily emitted electromagnetic signals produced by computer chips. This was possible because generative adversarial networks are learning machines that can be trained to produce plausible timeseries signal data from tokenized serial input like a text-to-speech synthesizer, and because photons are emitted whenever charged particles accelerate, such as when microcontrollers and microprocessors execute instructions. The attributes of the emitted photons form a unique and architecture specific signature for the instructions executed. The goal of this project was to produce an instruction-to- electromagnetic -waveform synthesizer that would enable accurate prediction of the involuntarily emitted electromagnetic signals produced by computer chips. To achieve this goal, a singular value decomposition-based framework for anomaly detection in noisy industrial environments was created, a database was constructed to map short sequences of instructions to their unique electromagnetic fingerprints, a metric was invented to compare synthesized electromagnetic signals to collected electromagnetic signals, and a novel learning machine pipeline was designed to create plausible electromagnetic signals for a computer chip from sequences of instructions.



The developed framework for training efficient electromagnetic based anomaly detection models on synthetic signals generated by a generative adversarial network.

TALENT PIPELINE:

- Kurt Vedros, student at University of Idaho
- Robert Ivans, student at Boise State University, converted to staff

PRESENTATIONS:

- E. Miller, G. M. Makrakis, K. A. Vedros, C. Kolias, C. Rieger, and D. Barbara, "Detecting Code Injections in Noise Environments Through EM Signal analysis and SVD Denoising," 20th International Conference on Embedded Systems, Cyber-physical Systems, and Applications (ESCS 22), Las Vegas, 2022.
- K. A. Vedros, C. Kolias, and R. C. Ivans, "Do Programs Dream of Electromagnetic Signals? Towards GAN-based Code-to-Signal Synthesis," MILCOM 2023-2023 IEEE Military Communications Conference (MILCOM), Washington, DC, 2023.
- K. A. Vedros, G. M. Makrakis, C. Kolias, R. C. Ivans, and C. Rieger, "Towards Scalable Anomaly Detection for Embedded Devices Through Synthetic EM Fingerprinting," 12th International Conference on Embedded Systems and Applications (EMSA2023), Vienna Austria, 2023.
- K. A. Vedros, C. Kolias, and R. C. Ivans, "From Code to EM Signals: A Generative Approach to Side Channel Analysis-based Anomaly Detection," International Conference on Availability, Reliability, and Security (ARES), Vienna Austria, 2024.

INTELLECTUAL PROPERTY:

- K. A. Vedros, R. C. Ivans, C. Kolias, and C. Rieger, "Systems and Methods for Synthetic Side-Channel Analysis," US application number 63/570,790, Mar. 2024.

Target-aware fuzzing

PROJECT NUMBER:
22A1059-129FP

TOTAL APPROVED AMOUNT:
$675,000 over 3 years

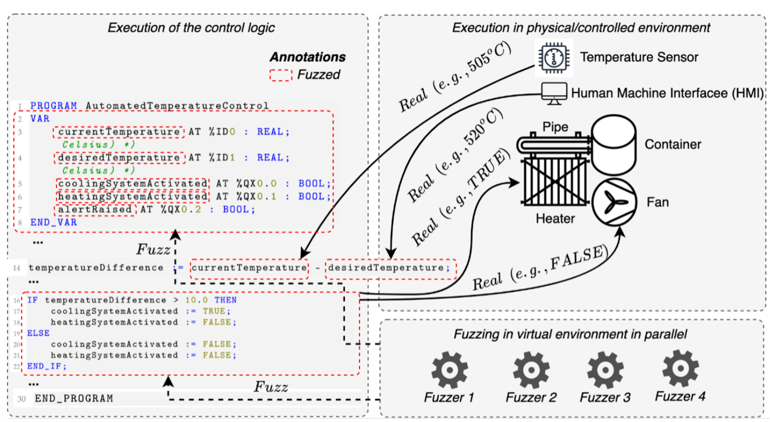
PRINCIPAL INVESTIGATOR:
Robert Ivans

CO-INVESTIGATORS:
Craig Rieger, INL
Michael McCarty, INL
Constantinos Kolias, University of Idaho

COLLABORATOR:
University of Idaho

A novel vulnerability discovery and reproduction framework finds bugs in critical infrastructure so they can be patched before adversaries have the chance to exploit them.

Programmable logic controllers are industrial computers that control critical infrastructure automated systems. The programs that programmable logic controllers run help automate electric power generation, transmission, and distribution, oil and gas production, and nuclear power plants. These programs are often written in specialized languages, such as Structured Text, described in International Electrotechnical Commission 61131-3. In software development, fuzzers are used to ensure that software is secure and reliable. Fuzzers are programs that rigorously test software by feeding them scrambled inputs to try and elicit unexpected behaviors or effects. Among the most popular fuzzers in the world is American Fuzzy Lop Plus Plus, which is an improved fork of Google's fuzzer American Fuzzy Lop. However, American Fuzzy Lop Plus Plus can not fuzz programs written in International Electrotechnical Commission 61131-3 languages. This project enables rigorous testing of industrial control system software written in Structured Text, and any International Electrotechnical Commission 61131-3 language that can be converted to Structured Text: Instruction List, Functional Block Diagram, and Ladder Logic. This was possible because of the fuzzing framework developed by this project, which integrates a custom programmable logic controller runtime and a purpose-built fuzzer. The goal of this project was to help keep American critical infrastructure safe by creating a fuzzing system capable of testing the industrial control system software that Americans depend on. To achieve this goal, a fuzzing framework specifically designed to target programmable logic controller software was developed, directional components were added to the fuzzing strategy, and a custom seed generation scheme was developed that enables user intuition to guide fuzzing.



Instance of the proposed framework for fuzzing control logic written in Structured Text.

TALENT PIPELINE:

- Koffi Anderson Koffi, student at University of Idaho
- Robert Ivans, student at Boise State University, converted to staff

PUBLICATIONS:

- K. A. Koffi, V. Kampourakis, J. Song, C. Kolias, and R. C. Ivans, "StructuredFuzzer: Fuzzing Structured Text-Based Control Logic Applications," Electronics, vol. 13, no. 13, p. 2475.
- K. A. Koffi, V. Kampourakis, J. Song, C. Kolias, and R. C. Ivans, "Speeding-up Fuzzing Through Directional Seeds," International Journal of Information Security, vol. 24, no. 77 (2025).

INTELLECTUAL PROPERTY:

- "Buggy program program," GitHub repository. GitHub, 2022. [Online]. Available: https://github.com/IdahoLabResearch/Buggy-Program-Program/
- "StructuredFuzzer," GitHub repository. GitHub, 2024. [Online]. Available: https://github.com/kandersonko/StructuredFuzzer

Artificial intelligence based confidentiality, integrity, and availability of wirelessly transmitted data in the nuclear industry

PROJECT NUMBER:
23A1070-127FP

TOTAL APPROVED AMOUNT:
$700,000 over 2 years

PRINCIPAL INVESTIGATOR:
Vivek Agarwal

CO-INVESTIGATORS:
Amitabh Mishra, INL
Joshua Daw, INL
Shannon Eggers, INL
Mingyue Ji, University of Utah
Senha Kumar Kasera, University of Utah

An innovative federated learning approach enables secure and reliable wireless communication in critical infrastructure.

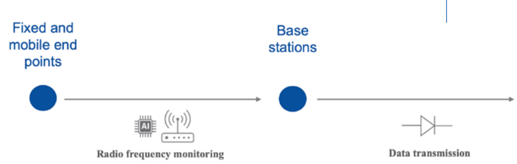
This project focused on improving the security and reliability of wireless communications in critical infrastructure such as nuclear power plants. Given the crucial role of sensor data in plant monitoring and operations, data transmission errors or anomalies of any kind can have serious consequences for plant safety and efficiency. Anomalies could be caused by sensor malfunction, data spoofing at the sensor level or the transmission level, or malicious cyberattacks. To address these concerns, the project developed an artificial intelligence based spectrum heatmap prediction and data anomaly detection algorithm along with robust statistical techniques to identify anomalies. A novel deep Gaussian process-based model was developed to create precise wireless heatmaps to identify unauthorized transmissions and detect anomalies in wireless environments, even with limited radio frequency data. Validation with real-world received signal strength data from the Platform for Open Wireless Data-driven Experimental Research (POWDER) at the University of Utah demonstrated that the developed deep Gaussian process model outperformed state-of-the-art standard Gaussian process models and deep neural network models. Furthermore, the developed deep Gaussian process model used federated learning, a distributed learning technique. Unlike centralized earning, federated learning allows each sensor node to process data locally, mapping the received signal strength data to various locations without the need to transmit the information to a centralized location. A novel federated Gaussian process model combined with Bayesian ensemble learning was developed to generate global model updates, which were compared to the simple averaging approach used in the literature. This approach significantly outperformed the existing federated Gaussian process methods and was validated using real-world data from POWDER. The project's success is a step toward achieving secure wireless communication in nuclear power plant operations. These outcomes have a cross-cutting impact across reactor technologies and other critical infrastructures. The advancements support the strategic goals of the INL Wireless Security Institute, representing a significant improvement in nuclear security and communication technology.

TALENT PIPELINE:

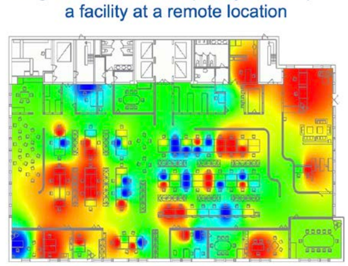
- Imtiaz Nasim, early career researcher at INL
- Xiang Zhang, student at University of Utah
- Yanyu Hu, student at University of Utah

PRESENTATIONS:

- X. Zhang, Y. Hu, A. Mishra, I. Nasim, J. E. Daw, S. L. Eggers, V. Agarwal, A. Bhuyan, S. K. Kasera, and M. Ji, "A Bayesian Learning Approach to Wireless Outdoor Heatmap Construction Using Deep Gaussian Process," The 58th Annual Asilomar Conference on Signals, Systems, and Computers, 2024.
- I. Nasim, V. Agarwal, J.E. Daw, M. Ji, and S. Kasera, "Statistical and Neural Network for Real Sensor-Data-Driven Anomaly Detection in Nuclear Applications," International Congress on Advances in Nuclear Power Plants (ICAPP), 2024.



AI generated radio frequency heatmap of a facility at a remote location



Demonstrating confidentiality, integrity, and availability of wireless transmitted data using artificial intelligence over University of Utah's POWDER. There are fixed and mobile end points that transmit the data wirelessly and have wireless monitors with artificial intelligence engines to the base station. From the base stations the data is transmitted to a remote monitoring center via a data diode for visualization (right).

Authentication protocol for wireless ad hoc networks with embedded certificates

PROJECT NUMBER:
23A1070-102FP

TOTAL APPROVED AMOUNT:
$461,000 over 2 years

PRINCIPAL INVESTIGATOR:
John Capson

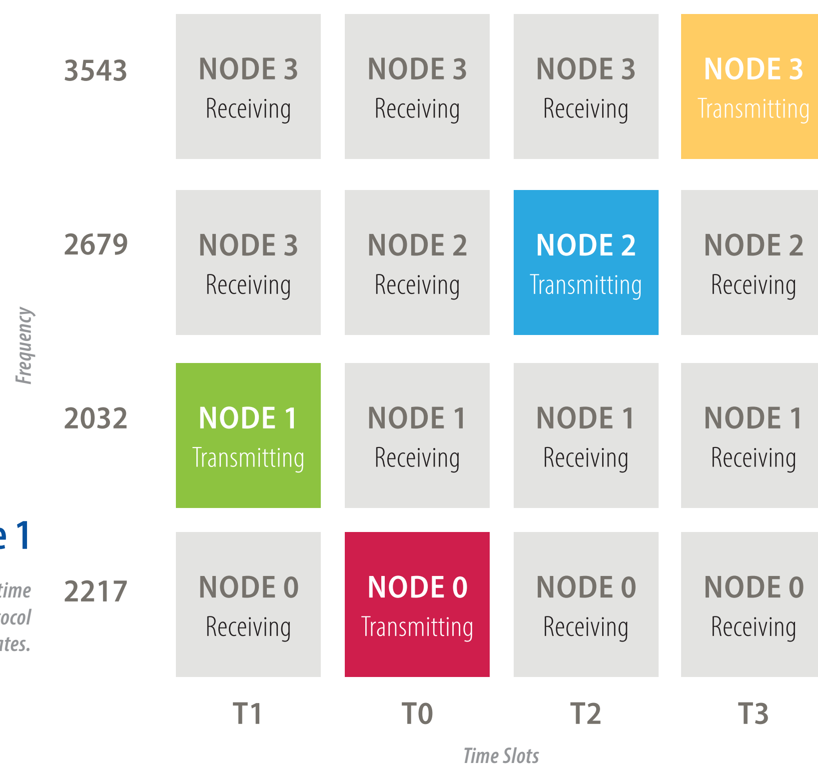
CO-INVESTIGATORS:
Andrew Weaver, INL
Robert Hiromoto, University of Idaho

A new authentication protocol for embedded certificates increases ad hoc network security.

Wireless ad hoc networks may be configured as a fixed topology of sensors or allowed to migrate as mobile nodes. This flexibility provides opportunities for their deployment in real-time and in adverse situations as encountered in civil and military applications. However, these advantages are curtailed by the unconstrained nature of these networks in providing a trusted level of connectivity. Establishing secret keys and authenticating trusted ad hoc group nodes are essential elements for a secure network. In this project, we developed an authentication protocol for wireless ad hoc networks that is derived from a canonical splitting of time- and frequency-space (channel) over which information propagates under the constraint of a collision-avoidance protocol. This authentication protocol for embedded certificates takes advantage of the physical communication traits, such as time slots and frequency slots, to embed the authentication into the communication stream automatically. This dramatically reduces the overhead compared to traditional certificate passing based authentication protocols. The protocol was implemented and verified in simulation and was then implemented on hardware using a software defined radio. It stood up to various attacks in simulation to include replay, jamming, and man in the middle attacks. The physical implementation withstood replay attacks with no protocol failures.

TALENT PIPELINE:

- Doug Hill, student at Brigham Young University, Idaho, converted to staff
- Spencer Saunders, student at Brigham Young University, Idaho converted to staff



Cycle 1

Division of frequency and time slots using authentication protocol for embedded certificates.

Quantifying organizational influence on critical infrastructure systems

PROJECT NUMBER:
23A1070-133FP

TOTAL APPROVED AMOUNT:
$457,800 over 2 years

PRINCIPAL INVESTIGATOR:
Gabriel Weaver

CO-INVESTIGATOR:
Daniel Eisenberg,
Naval Postgraduate School

COLLABORATOR:
Orca AI

New software platform simplifies and accelerates the identification of infrastructure threats enabled by organization and business relationships.

This project developed analytic techniques to identify collections of sociotechnical dependencies that create conditions conducive to adversarial behavior. Although much research focuses on cyber-physical dependency models and risk analysis, an entire class of threat models enabled via legal business practices has the potential to impact critical infrastructure components across their entire life cycle. Such analysis addresses key problems faced by DOE and the US, including cyber supply chain risk management and characterization of risks associated with the clean energy transition. For example, within the energy sector, the trend toward decentralization of ownership, operations, and maintenance of facilities, results in more complex business relationships that increase operational risk. The Office of the Director of National Intelligence recently noted that venture capital investment has become a "permissive target for foreign investors, including state actors, seeking to gain a foothold into the supply chain of a new technology or an area of national security or strategic interest." DOE needs more efficient, consistent, and scalable analytic capabilities in this domain.

Therefore, we developed the Technology, Organization, and Person of interest Graph Extraction, Analysis and Reporting (TOPGEAR) platform tailored to assess soft-power relationships and their impacts on critical infrastructure. Current approaches in the field and industry are largely manual, limited in scale, and time-consuming. TOPGEAR is a laboratory-grade software tool that is available at the INL Software Store. TOPGEAR's primary function is to simplify the ability to iteratively crawl business relationships across multiple data sources, fuse them together, analyze indicators for soft-power influence, and present results for further investigation. Our research has been presented via conferences, peer reviewed publications, and a Master's thesis. Specifically, the work has focused on case studies involving regional electric vehicle charging stations, though the capability has a variety of application domains.

The project's harvest strategy resulted in a presentation of the capability at INL's Strategic Advisory Council as well as proposals to DOE's Office of Cybersecurity, Energy Security, and Emergency Response, DOE's Office of Clean Energy Demonstrations, and the Department of Homeland Security National Risk Management Center.

TALENT PIPELINE:

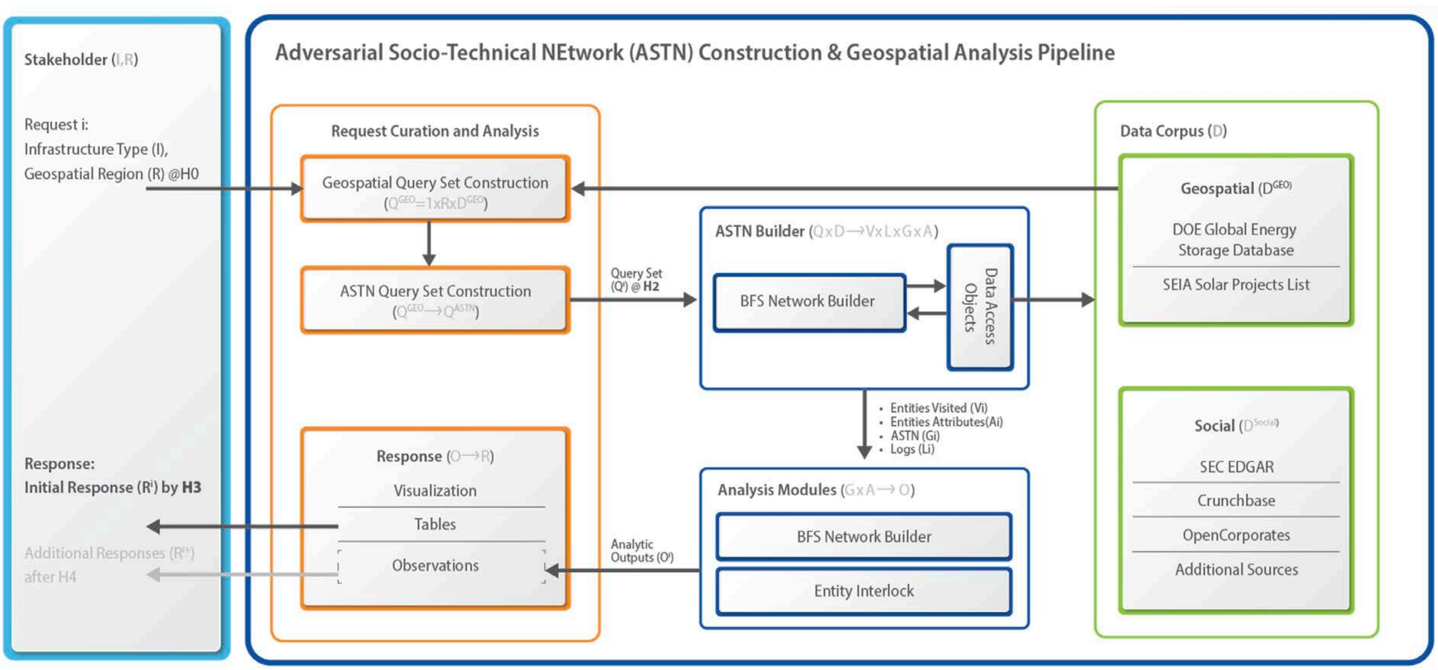
- Ryan Shannon, student at Naval Postgraduate School

PRESENTATIONS:

- Gabriel A. Weaver, Daniel A. Eisenberg, "Comparison of Sociotechnical Threat Models," Military Operations Research Society Symposium, US Military Academy, West Point, NY, 2023.
- Gabriel A. Weaver, Daniel A. Eisenberg, "A Data Processing Pipeline for Sociotechnical Network Analysis," Military Operations Research Society Symposium, US Military Academy, West Point, NY, 2023.

INTELLECTUAL PROPERTY:

- TOPGEAR: Technology, Organization, and Person of interest Graph Extraction, Analysis, and Reporting, https://inlsoftware.inl.gov/product/topgear



The TOP GEAR workflow consists of a stakeholder request, analyst query curation, and multilayered network construction and analysis. Generated networks are annotated with primary sources from which relations were derived.

Reinforcement learning based approach to optimizing quality of service and security on fifth generation networks

PROJECT NUMBER:
23A1070-134FP

TOTAL APPROVED AMOUNT:
$740,000 over 2 years

PRINCIPAL INVESTIGATOR:
Cameron Krome

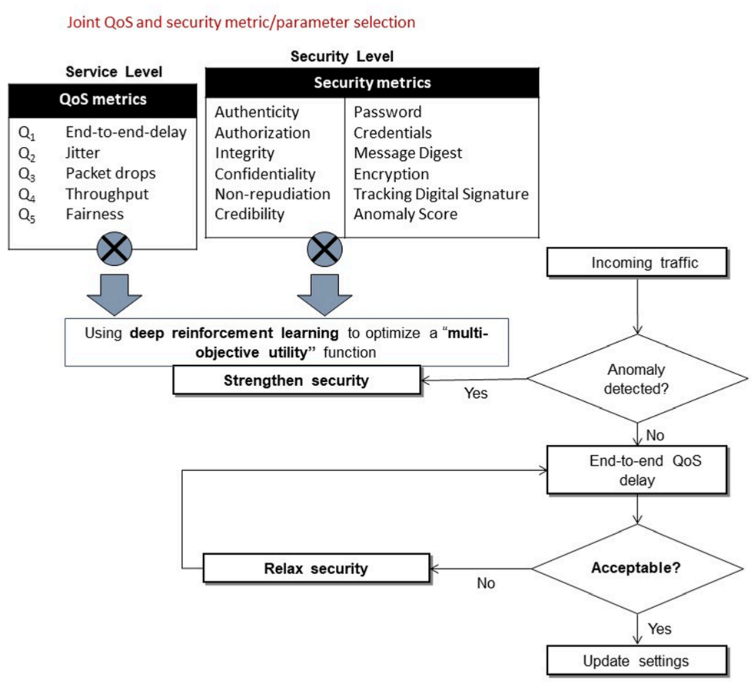
CO-INVESTIGATORS:
Anna Quach, INL
Christopher Becker, INL
Israel Olaveson, INL
Jared Wadsworth, INL
Shad Staples, INL
Mostafa Fouda, Idaho State University

COLLABORATOR:
Idaho State University

Actively adjusting Wi-Fi and fifth generation configuration settings improves quality of service.

In the evolving landscape of digital communication, the surge in wireless network utilization has become a prominent trend, bringing with it a pressing challenge: as network saturation increases, the quality of service diminishes. This service degradation is a consequence of increased traffic and is exacerbated by the enhanced security measures essential to safeguarding the network. The inverse relationship between heightened security protocols and quality of service underscores the necessity for innovative strategies that bolster service quality without compromising security. To address this issue, researchers at INL created two distinct wireless networks—one Wi-Fi based and the other a simulated fifth generation environment. These networks were used to generate data for use in further evaluating the problem and later training machine learning models.

The resulting datasets were analyzed to deepen the understanding of how quality of service and security configuration interact. The data was then used to train a variety of reinforcement learning models of varying complexity. The objective of the models was to maximize the quality of service by making changes to the wireless network configuration on a moment-to-moment basis. The models operate under a pivotal constraint: any adjustments proposed to improve service quality must not undermine the network's security posture. Through iterative learning and adaptation, these models identified and executed configuration changes that strike a delicate balance between robust security and high quality service delivery.



Proposed joint optimization of quality of service and security service parameters.

TALENT PIPELINE:

- Hamza Kaddour, student at Idaho State University
- Israel Olaveson, student at Brigham Young University–Idaho
- Mostafa Fouda, postdoc at Idaho State University
- Thomas Kopcho, student at Idaho State University

PRESENTATIONS:

- Kopcho, T., Jr., Fouda, M., Krome, C., Quach, A., Olaveson, I., "Analyzing the Impact of Security Measures on Wi-Fi Quality of Service," 2nd International Conference on Artificial Intelligence, Blockchain, and Internet of Things (AIBThings), September 7 – 8, 2024.
- Kopcho, T., Jr., Fouda, M., Krome, C., A Lightweight AI Model for Anomaly Detection in Wireless Networks, 2nd International Conference on Artificial Intelligence, Blockchain, and Internet of Things (AIBThings), September 7 – 8, 2024.

INTELLECTUAL PROPERTY:

- Krome, C., Olaveson, I., Kopcho, T., Jr., Quach, A., "Idaho National Laboratory Quality of Service Dataset," https://github.com/Iolaveson/QoS_Idrd

Seismic resilience of transformers

PROJECT NUMBER:
23P1075-018FP

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PRINCIPAL INVESTIGATOR:
Bjorn Vaagensmith

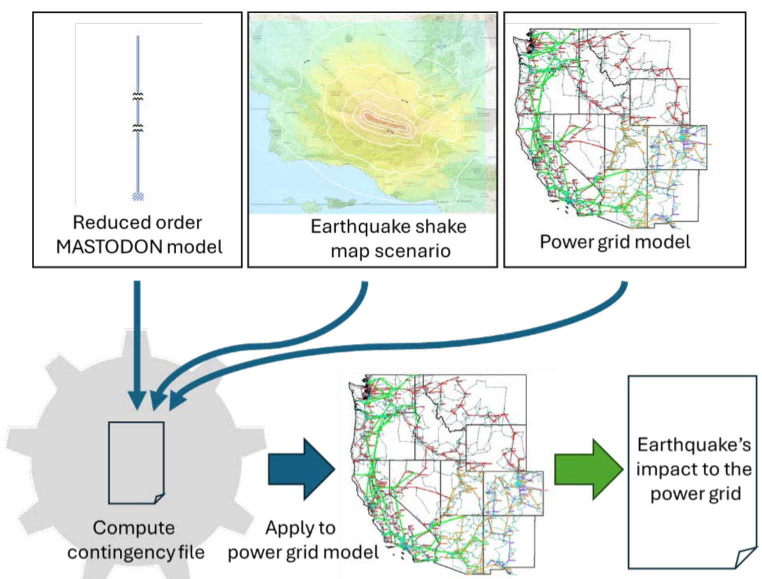
CO-INVESTIGATORS:
Chandrakanth Bolisetti, INL
Jon Bender, W. E. Gundy and Associates

Modeling transformers helps planners mitigate grid failures due to earthquakes.

A large earthquake in a population dense region in the Western US has the potential to result in a blackout for millions of people for multiple months due to a critical vulnerability—failure of power transformer bushings due to vibrational coupling with transformer tanks. Western US hosts about a quarter of the US population and experiences major seismic activity. A single failed transformer can cost tens of millions of dollars and require up to five years lead time due to an imbalance in global manufacturing capability versus demand. Despite this known seismic vulnerability of the transformers, its impact on the grid still needed to be quantified. To solve this problem, this project developed a methodology that combined dynamic finite element modeling of transformers with power grid modeling. Dynamic finite element modeling was used to generate risk profiles of various transformers by voltage class. These risk profiles were used to generate thousands of contingency cases based on various earthquake scenarios from the power grid.

TALENT PIPELINE:

- Akram Batikh, student at North Carolina State University
- Alex Harvey, student at North Carolina State University
- Hasan Khan, student at University of Texas at Austin
- Jet Wo, student at University of Texas at San Antonio
- Joseph Liebergen, student at Dakota State University



Graphic showing the general process of the grid impact assessment.

High performance Boolean satisfiability computing

PROJECT NUMBER:
23P1080-017FP

TOTAL APPROVED AMOUNT:
$145,000 over 1 year

PRINCIPAL INVESTIGATOR:
Lance Joneckis

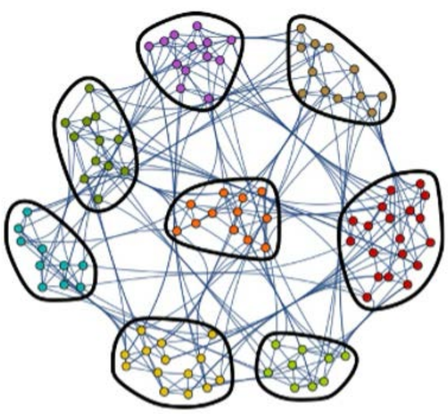
CO-INVESTIGATOR:
Gregory Shannon, INL

Designing non-traditional computing architectures with correspondingly new algorithms may improve Boolean satisfiability computations by a factor of 10 or more as compared to current parallelization methods.

This research evaluated the feasibility of using codesign—non-traditional computing architectures with correspondingly new algorithms—to improve the parallelizability of Boolean satisfiability computations by a factor of 100 or more compared to current methods that achieve approximately a factor of 10 speedup by parallelization. State-of-the-art satisfiability computations are not scalable to very large, hard problems. The best algorithms work on a single central processing unit core with parallel approaches using multiple cores demonstrating asymptotically negligible to negative speedup. This research was a first step in using codesign of algorithms and hardware to increase the computational performance for hard or large satisfiability instances by a factor of 100 to 1,000. Our effort at this stage concerns the conception and preliminary technical analysis of novel computational paradigms for parallel satisfiability computations based on codesign to achieve scalability improvements.

TALENT PIPELINE:

- Sara Logsdon, student at University of Georgia



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9	25	5	5	3	3	6	3
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6	3	4	7	21	4	4	3
7	3	4	5	4	21	3	4
2	6	5	2	4	3	18	2
5	3	2	5	3	4	2	15

CLUSTER-BLOCKED ADJACENCY MATRIX

CLUSTER-BLOCKED ADJACENCY PLOT

Illustration of the decomposition of a large Boolean satisfiability instance into smaller clusters. The clusters are determined by community structure where there are more internal links in a cluster than there are links connecting clusters. The decomposition allows for large Boolean satisfiability instances to be distributed over multiple computational elements.

CHEMICAL AND MOLECULAR SCIENCE

CORE CAPABILITIES

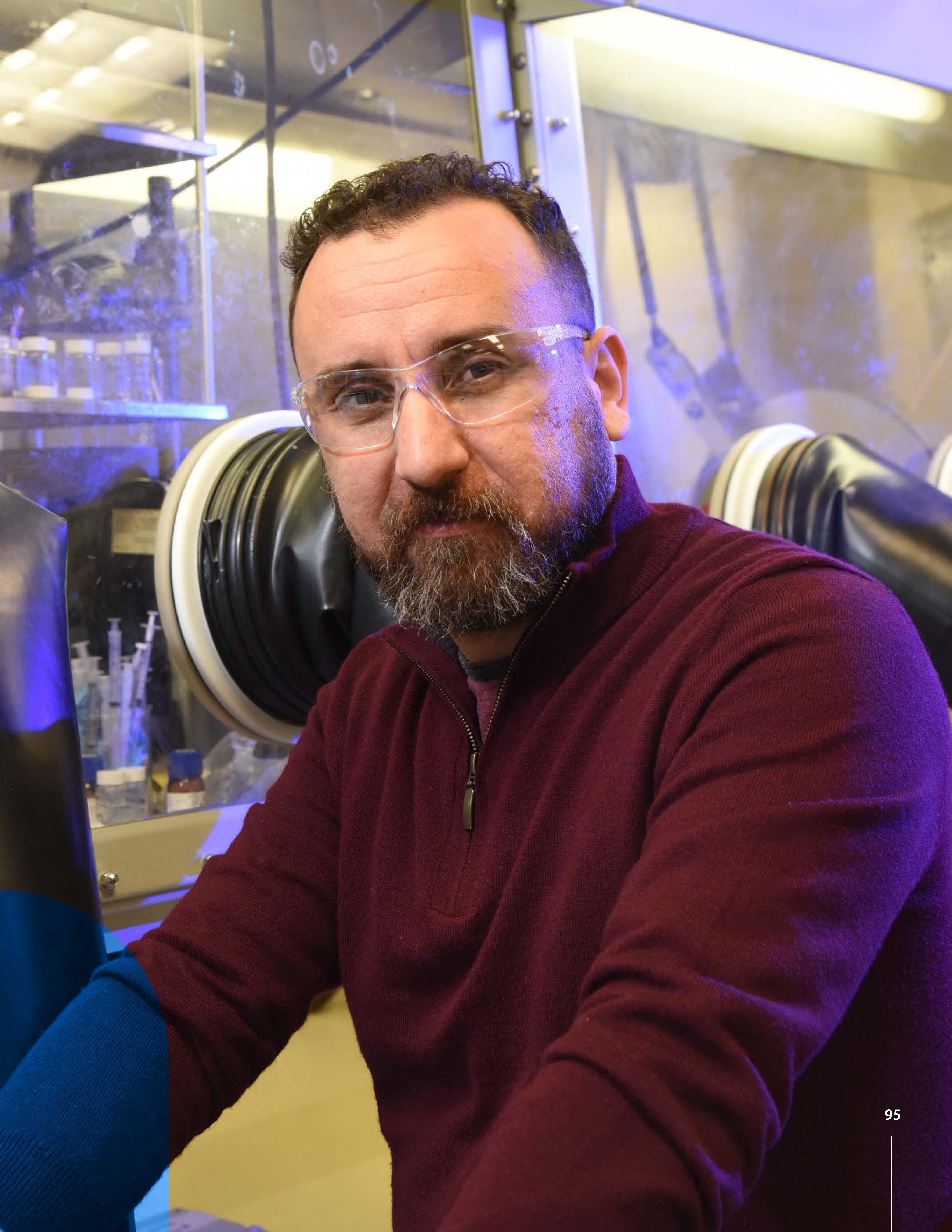
Chemical and Molecular Science

Applied Materials Science & Engineering

Computational Science

INL's chemical and molecular science capability explores fundamental research in nuclear science and radiation and actinide chemistry, and addresses important challenges in low carbon energy, energy conversion and storage, national security, and the environment. Our scientists use this capability to advance basic research in energy delivery systems, advanced and critical materials, clean water, renewable chemicals, transient kinetic analysis, and chemical separations. Of the 11 projects in this report that support chemical and molecular science, this core capability was the primary focus of the six projects in this section.

ABDERRAHMAN ATIFI



Intrinsic reactivity of energy production materials

PROJECT NUMBER:
22A1059-125FP

TOTAL APPROVED AMOUNT:
$789,000 over 3 years

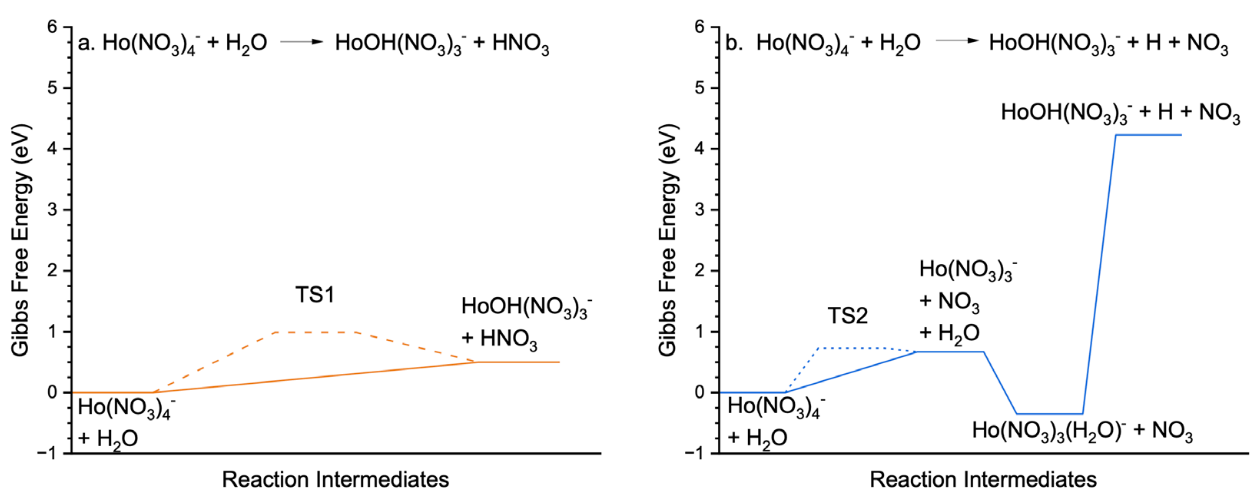
PRINCIPAL INVESTIGATOR:
Christopher Zarzana

CO-INVESTIGATORS:
Brittany Hodges, INL
JungSoo Kim, INL
Meng Li, INL

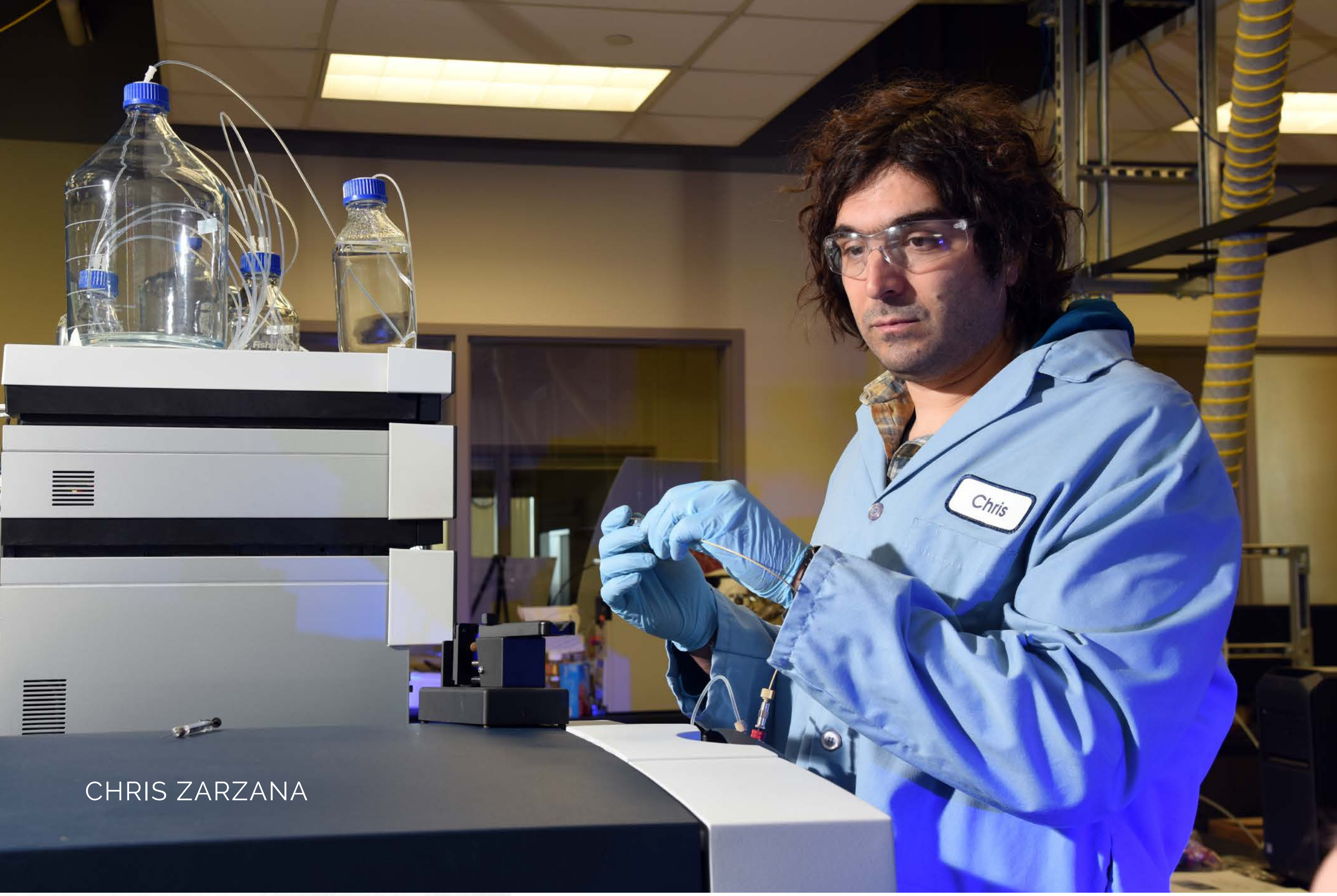
COLLABORATOR:
Université de Lorraine

Coupling gas-phase ion chemistry experiments with computational electronic structure theory provides insight into the water splitting mechanism on lanthanide-containing complexes.

Efficient energy storage, transport, and conversion is critical to developing a low-carbon economy. Reversible solid oxide cells that can either convert electricity to chemical energy or chemical energy back to electricity will be an important component of the energy production, storage, and distribution networks required to power a low-carbon energy future. Efficiency improvements of these devices rely on developing novel materials with enhanced performance; however, research in this area is slow due to the large number of possible material compositions and an underdeveloped understanding of the device performance mechanisms due to challenges interrogating the fundamental chemical reactions involved. New capabilities are needed to accelerate development of the materials that will allow reversible solid oxide cells to reach their full potential. This project studied the influence of electronic structure on water splitting reactions, which will help guide development of new materials used to convert water to hydrogen using electricity. Water splitting has been observed for gas-phase nitrate complexes derived from some—but not all—of the lanthanides. Thus, investigation of water interaction with gas-phase lanthanide nitrate complexes enables explorations of how small changes in electronic structure can enable water splitting reactions. Details of the chemical mechanism for this water splitting reaction are required to begin to understand how electronic structure influences outcomes. Our experiments demonstrated that the previously hypothesized splitting mechanism is unlikely. This research developed two alternate reaction mechanisms, which we have investigated with electronic structure theory calculations. Improved understanding of chemical reactivity will support rapid materials screening implementation that will accelerate transformative materials development for chemical and electrical energy interconversion, enabling deployment of the efficient energy storage, transport, and conversion networks required for a low-carbon economy.



Potential energy surface calculations for two possible mechanisms for the gas-phase splitting of water on holmium nitrate complexes to form holmium hydroxide trinitrate ([HoOH(NO3)3]-).



CHRIS ZARZANA

TALENT PIPELINE:

- JungSoo Kim, postdoc at INL

PRESENTATION:

- Kim, J., Zarzana, C., Li, M., Hodges, B., "The investigation of the mechanism for the water splitting by holmium oxide nitrate complex in gas-phase," 71st American Society for Mass Spectrometry (ASMS) conference on Mass Spectrometry and Allied Topics, June 4th-8th, 2023, Huston, TX, USA.

Metaheuristic machine learning accelerated quantum chemistry for investigating multiphase interactions in electrochemical systems

PROJECT NUMBER:
23A1070-027FP

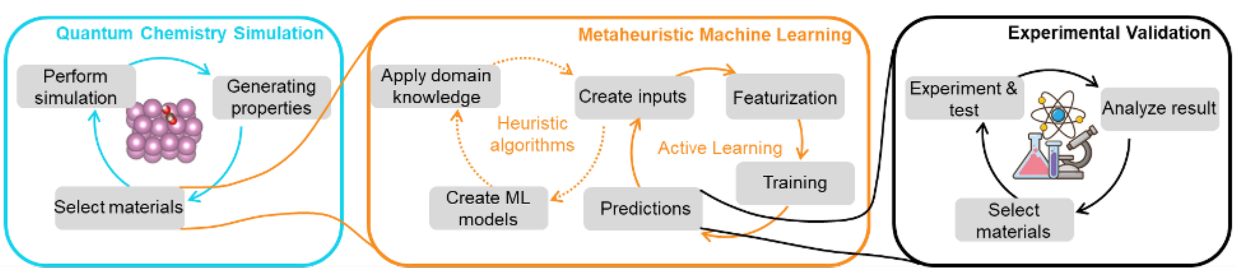
TOTAL APPROVED AMOUNT:
$806,000 over 2 years

PRINCIPAL INVESTIGATOR:
Meng Li

CO-INVESTIGATORS:
Boryann Liaw, INL
Dong Ding, INL
Wenjuan Bian, INL
Leslie Kerby, Idaho State University
Collaborators:
Kansas State University
Massachusetts Institute of Technology
Montana State University

Machine learning accelerated quantum chemistry framework enables efficient electrocatalyst development in a vast material space.

Hydrogen is an important energy carrier resource that helps limit greenhouse gas emissions. Solid oxide electrolysis cells, including proton-conducting and oxygen-conducting electrolyzers, are highly efficient carbon-neutral hydrogen technologies. The features central to their systems are a multitude of multiphase interactions at interfaces that influence driving forces for both desired and undesired reactions. The complexity of multiphase interactions provides numerous grand challenges in terms of understanding the detailed properties for performance improvement and materials development. Therefore, new approaches are essential to developing efficient electrocatalysts for selective reactions for target products among the vast material composition space. High entropy alloys exhibit fascinating applications for hydrogen evolution reaction, showcasing their versatility in promoting clean energy technologies. The extensive compositional diversity inherent in high entropy alloys has given rise to numerous unique properties. However, the size of this compositional space poses a formidable challenge for both comprehensive modeling and experimentation. Given the complexity of high entropy alloys, it becomes impractical to explore the entire compositional space, especially when considering a simple high entropy alloy comprising 100 elements with 20 atoms of each element. The number of possible combinations in such a scenario reaches a staggering 105 possibilities, making it impossible to investigate the entire space. This project developed a robust predictive framework to understand multiphase interactions at solid/gas interfaces and predict surface activity by coupling machine learning and quantum chemistry. The predictive framework goes beyond common composition/structure-property-behavior relationship identification. Its transferability between systems and processes will facilitate the development of more efficient systems for energy storage, transport, and conversion.



Metaheuristic machine learning accelerated density functional theory calculation workflow for developing efficient electrocatalysts.

TALENT PIPELINE:

- Alexander Svancara, student at Idaho State University
- Daniel Igbokwe, student at Idaho State University
- Dunya Bahar, student at Idaho State University
- Grant Madson, student at Idaho State University
- Saugat Acharya, student at Idaho State University
- Sharmistha Das Karmakar at postdoc, INL

PRESENTATIONS:

- M. Li, D. Dong, "Accelerated Discovery of Proton-Conducting Perovskites through Density Functional Theory and Machine Learning," 242nd ECS Meeting, October 9-13, 2023.
- M. Li, B. Hua, L.-C. Wang, W. Wu, D. Ding, "Tuning Selective CO2 Electrohydrogenation Under Mild Temperature and Pressure," Invited presentation at the 245th Electrochemical Society Meeting, May 26–30, 2024.
- M. Li, "Experiments and AI Converge: Decoding Physical and Chemical Principles," Invited keynote presentation at the Symposium on Data, AI and Materials Analytics at Corning Incorporated, September 18, 2024.

Identifying the speciation of salt-based actinides in the presence of contaminants to provide insight into heat transfer of molten salt reactors

PROJECT NUMBER:
23P1077-002FP

TOTAL APPROVED AMOUNT:
$446,000 over 2 years

PRINCIPAL INVESTIGATOR:
Liyanage Ashini Jayasinghe

CO-INVESTIGATORS:
Aaron Wilson, INL
Julie Bowen, INL

Lanthanide and actinide chlorides in molten salts react with oxygen to form oxides and oxychlorides.

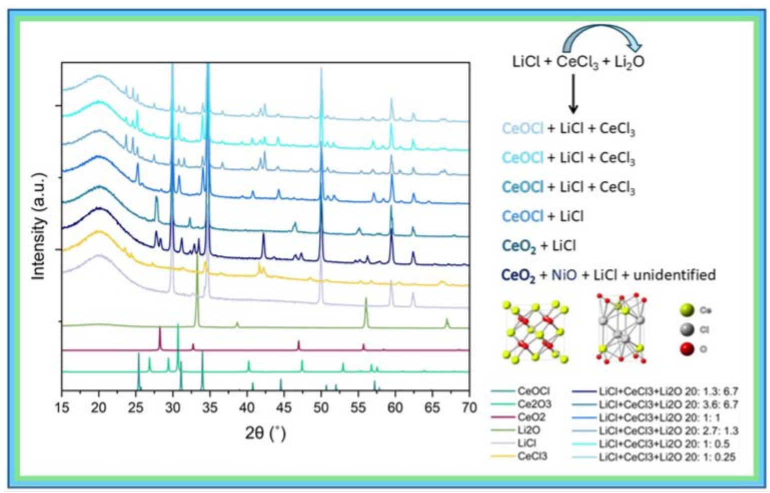
Advances in nuclear energy technology are vital for achieving net-zero emissions by 2050, and molten salt reactors are among the most promising Generation IV nuclear reactors. The salts used in these reactors exhibit a high affinity for moisture and oxygen, and heating does not eliminate the oxides that are formed. Additionally, due to the high operating temperatures, reactor parts experience pronounced corrosion, and the resulting corrosion products integrate into the fuel salt system. It was hypothesized that the speciation of f-elements in molten salts would change in the presence of oxides and corrosion products. These changes in local structure can alter the rheological properties, which are critical to the operational and safety parameters of the reactors. Therefore, an in-depth understanding of how corrosion products influence lanthanide/actinide speciation is crucial. To test this hypothesis, lanthanides (as surrogates for actinides) and actinide chlorides were mixed with oxides (such as nickel oxide and lithium oxide) and corrosion products (e.g., nickel chloride) and reacted in molten salts. The resulting phases were analyzed using powder x-ray diffraction, Raman spectroscopy, ultraviolet-visible spectroscopy, and nuclear magnetic resonance spectroscopy. Due to the high oxophilicity of f-elements, these elements capture oxygen from the oxides, forming lanthanide/actinide oxide or oxychloride phases. With cerium chloride ($CeCl_3$), introducing lithium oxide (Li_2O) resulted in various final products depending on the oxide concentration. At lower oxide concentrations, cerium oxychloride (CeOCl) in the +3 oxidation state was observed, while an increase in the oxide concentration led to the formation of cerium dioxide (CeO_2) where cerium is in the +4 oxidation state. In contrast, with nickel oxide (NiO), CeOCl formation was consistently observed regardless of the oxide concentration. Researchers determined that the speciation of f-elements changes in the presence of oxides and different oxides in molten salts result in different final products. When the influence of nickel chloride ($NiCl_2$) was examined, changes in f-element speciation were not observed. However, $NiCl_2$ reacted with lithium chloride to generate a lithium-nickel-chloride (Li_6NiCl_8) phase, which was also observed when nickel oxide was reacted. Computational models were developed to simulate the phases anticipated under experimental conditions. Collaborations were established with Brookhaven National Laboratory to access their beamline facilities for characterizing the generated salt samples, and additional internal collaborations were established to computationally model the reactions occurring in molten salts.

TALENT PIPELINE:

- Liyanage Ashini Jayasinghe, *Glenn T. Seaborg distinguished postdoctoral fellow at INL*, converted to staff

PRESENTATIONS:

- Jayasinghe, A., Bowen, J., Wilson, A., Gakhar, R., "Exploring the impact of molten-salt reactor contaminants on actinide speciation," Invited presentation at TMS annual Meeting, Orlando, FL, 04 March 2024.
- Jayasinghe, A., Bowen, J., Wilson, A., Gakhar, R., "Identifying the speciation of salt-based lanthanides in the presence of molten-salt reactor contaminants," ACS National meeting, San Francisco, CA, 14 August 2023.
- Jayasinghe, A., Bowen, J., Wilson, A., Gakhar, R., "Exploring the speciation of actinide salts in the presence of contaminants related to molten salts reactors," Actinides 2023, Golden, CO, 06 June 2023.



Cerium reacts with Li_2O in molten salt system due to cerium's oxophilicity. Powder x-ray diffraction data demonstrated that at lower Li_2O concentrations CeOCl was formed, CeO_2 was generated at higher Li_2O concentrations.

Investigating the impact of donor atoms of single-source precursors on f-element nanomaterial formation and mechanisms of initial bond cleavage

PROJECT NUMBER:
23P1080-025FP

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PRINCIPAL INVESTIGATOR:
Liyanage Ashini Jayasinghe

CO-INVESTIGATORS:
Christopher Zarzana, INL
Andrew Holland, Idaho State University
Joshua Pak, Idaho State University

Synthesizing and studying novel ligands and cerium compounds leads to a mechanistic understanding of cerium oxide nanoparticle formation from single-source precursors.

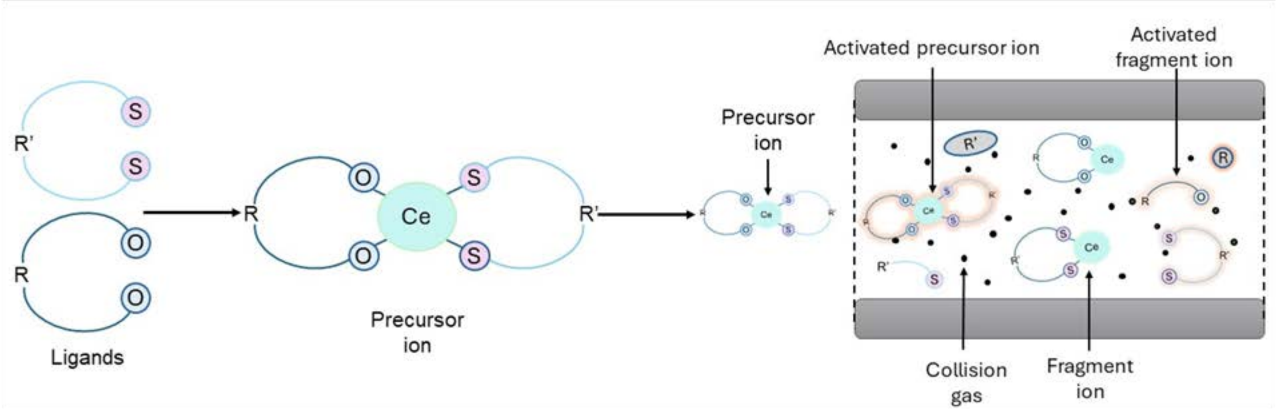
Nanotechnology is advancing exponentially with novel discoveries and applications emerging daily across various fields, including environmental remediation, biomedicine, electronics, and textiles. Nanoparticles are defined as particles with sizes ranging from 1 to 100 nm, and their properties are influenced by factors such as composition, phase, size, and shape. Single-source precursors offer a promising method to control phase-pure nanoparticle synthesis with the desired stoichiometry and have been successfully applied to prepare transition metal and lanthanide nanoparticles. Single-source precursors are molecular compounds that contain all the necessary components to form the final nanomaterial and decompose under mild conditions, such as heating, to create the solid state product. This research developed a mechanistic understanding of cerium oxide nanoparticles formation from cerium single-source precursors. First, known and novel ligands with oxygen and sulfur donor atoms were synthesized. These ligands were then chelated to cerium ions to synthesize cerium precursors. Initially, known cerium alkoxide precursors were synthesized. Subsequently, these basic cerium complexes were further manipulated through acid-base protonolysis reactions to install a diverse set of ligands. Within this body of work, ten novel cerium precursors were synthesized. The ligands and the single-source precursors were analyzed using nuclear magnetic resonance and thermogravimetric analysis coupled with differential scanning calorimetry. These single-source precursors were then introduced to a collision-induced dissociation mass spectrometer, and their dissociation pathways were studied. Early career researchers, including undergraduates, graduate students, and postdoctoral researchers, were actively involved in this project.

TALENT PIPELINE:

- Ainsley Snyder, student at Idaho State University
- Jonathan Pomeroy, student at Idaho State University
- JungSoo Kim, postdoc at INL
- Kameron Bowers, student at Idaho State University
- Liyanage Ashini Jayasinghe, *Glenn T. Seaborg distinguished postdoctoral fellow at INL*, converted to staff
- Mikaela Spafford, student at Idaho State University

PRESENTATION:

- Snyder, A., Spafford, M. L., Bowers, K., Pomeroy, J., Holland, A., Pak, J., Zarzana, C., Jayasinghe, A., JungSoo, K., "Synthesis of cerium precursors with alkoxide ligands for degradation to cerium oxide nanoparticles," ACS National meeting, Denver, CO, August 2024.



Ligands with oxygen and sulfur donor atoms were synthesized and chelated to cerium to form single-source precursors. The decomposition of these were explored using collision-induced dissociation mass spectrometry.

Revealing small ion intercalation dynamics through data mining of trajectories

PROJECT NUMBER:
24A1083-009FP

TOTAL APPROVED AMOUNT:
$200,000 over 1 year

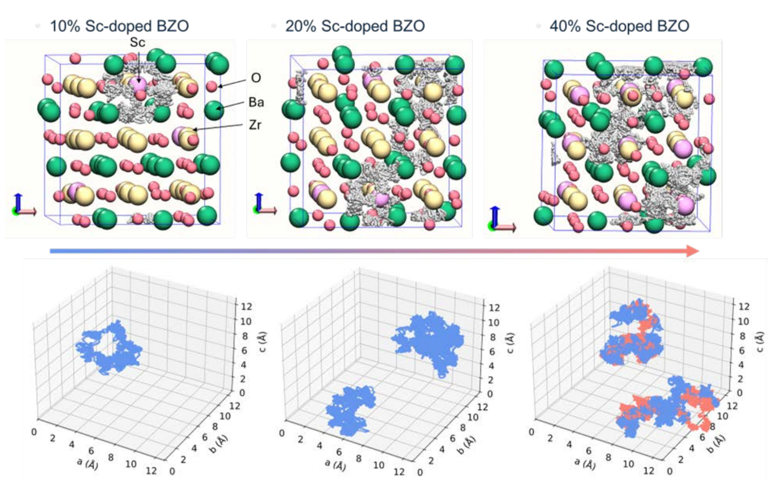
PRINCIPAL INVESTIGATOR:
Meng Li

MENTORS:
Yanwen Zhang
Krzysztof Gofryk

COLLABORATOR:
Kansas State University

A data mining framework to analyze small ion intercalation dynamics at the atomic level provides profound insights into ion behavior in host materials.

Ion intercalation, especially with small ions, is foundational to modern technologies like energy storage and conversion, physical neural networks in machine learning devices, and more. This process, which involves the insertion and removal of ions into a host material without substantial structural changes, is crucial for optimizing the efficiency, durability, and safety of systems. Typically, researchers harness the capabilities of density functional theory and molecular dynamics simulations to attain fundamental understandings. Yet, the derived ion migration barriers and diffusivities are only average measures that inadequately elucidate complex intercalation mechanisms at the atomic level, particularly in materials with complex structures and multiple dopants. Such approaches tend to overlook atomic dynamics because analyzing the vast simulation datasets remains a daunting task. Macroscopic ion conductivity and stability of host materials are controlled by microscopic ion motion in their microstructure spaces. This project developed a data mining framework tailored to analyze small ion trajectories within host materials, thereby furnishing comprehensive dynamic insights into the examined system. The focus for this study was on the proton intercalation process in a doped barium zirconate (BZO) lattice. The proton intercalation in the host lattice can be adjusted through the hydration process, leading to different protonation levels. The changes in local environments, such as an increased number of neighboring proton atoms, greatly influence proton movement. This research linked patterns of proton movement with insights derived from experimental observations by extracting meaningful information from ion trajectories. It crossed the gap between atomic and electronic attributes of materials and their macroscopic properties. Beyond elucidating proton intercalation dynamics in inorganic materials, this analysis method offers the potential to reveal other small ion dynamics. Moreover, it will pave the way for sophisticated data mining frameworks tailored to analyze ion trajectories, thereby furnishing comprehensive dynamic insights in various systems.



Proton movements in the scandium (Sc) doped BZO lattice at various doping concentrations at 900 K and the corresponding proton trajectory analysis using data mining.

PRESENTATIONS:

- M. Li, "Beyond Composition: The Effects of Local Reaction Environment on Activity, Selectivity, and Stability," Invited presentation at Telluride Science's workshop of High Temperature Energy Conversion: Electrochemical Oxidation and Reduction Mechanisms, June 15-19, 2024.
- M. Li, D. Ding, Y. Zhang, "Revealing Small Ion Intercalation Dynamics through Data Mining of Trajectories," The 24th International Conference on Solid State Ionics, July 15-19, 2024.

AWARD:

- Meng Li, 2024 Asian American Most Promising Engineer of the Year Award, August 21, 2024.

Rationalizing the role of f-orbitals coordination in the rare earth metals electrodeposition in organic electrolytes

PROJECT NUMBER:
24A1083-010FP

TOTAL APPROVED AMOUNT:
$200,000 over 1 year

PRINCIPAL INVESTIGATOR:
Abderrahman Atifi

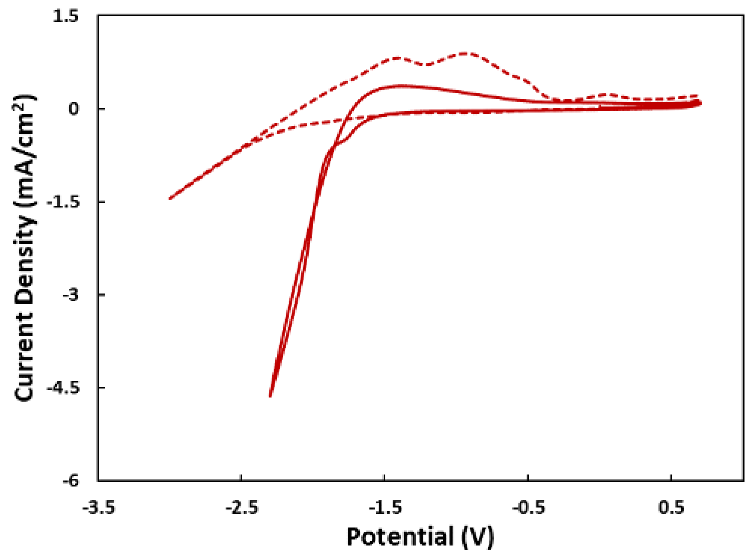
MENTOR:
Simon M. Pimblott

New research advances the fundamental understanding of rare earth metal solvation in electrolyte conditions.

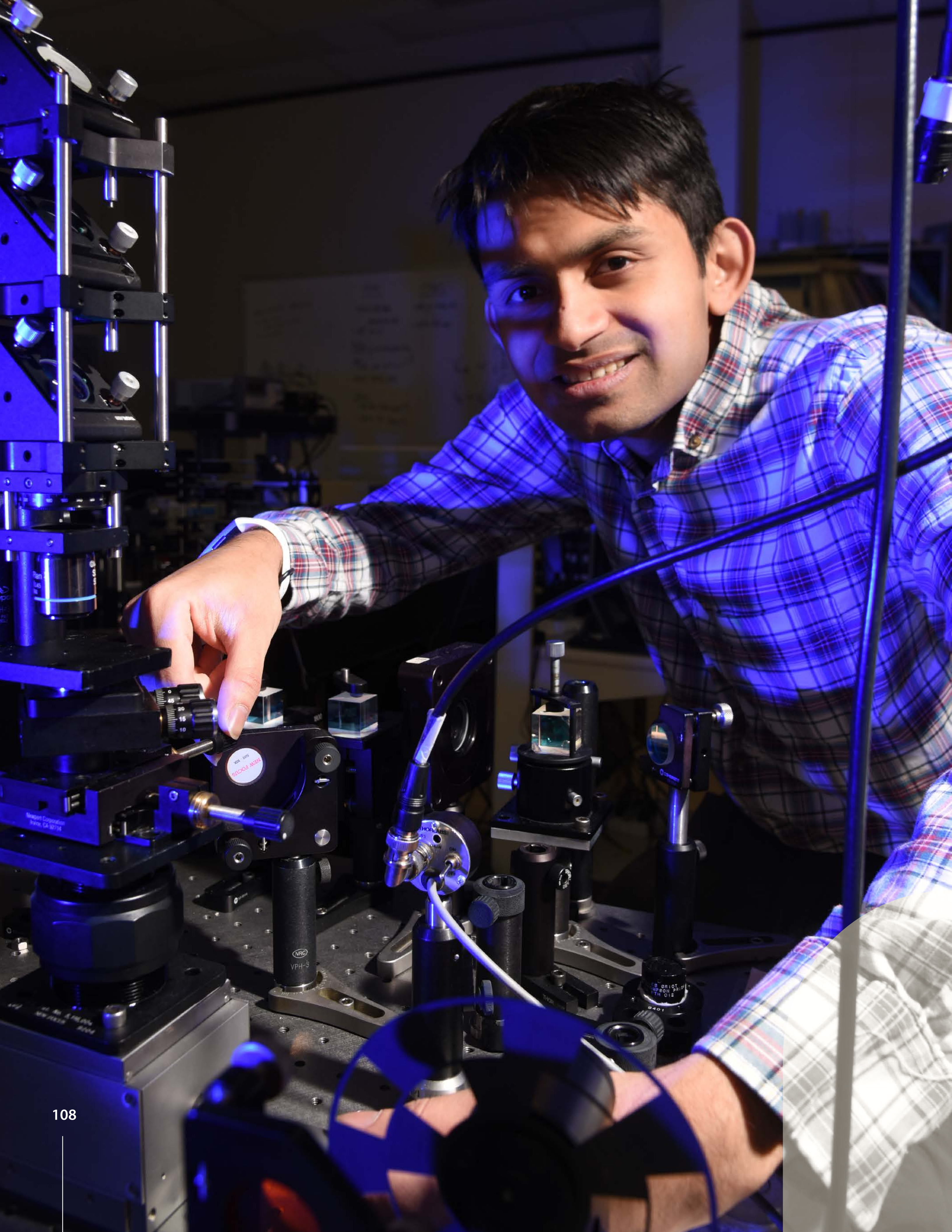
While the chemical bonding characteristics of f-elements remain a subject of debate in contemporary inorganic and coordination chemistry, their potential impact on electrochemical behavior has yet to be explored. The electrodeposition of rare earth metals in organic electrolytes has been shown to exhibit an irreversible reduction process. Borohydride-based electrolytes have emerged as promising media for the rare earth metal electrodeposition. We investigated the rare earth coordination and bonding nature in supported and concentrated electrolyte conditions. Our approach involved several analytical characterizations, including electrochemical analyses, electronic and vibrational configurations, nuclear magnetic resonance and quantum chemical calculations. We determined that the rare earth coordination environment is significantly influenced by the reaction mechanism, which is primarily controlled by stochiometric factors. Contrary to non-conductive and diluted media, concentrated neodymium dissolution in the supported electrolyte is primarily driven through an addition reaction. Under these reaction conditions, the dominant rare earth speciation exhibits a mixed coordination ligand sphere, featuring a low coordination number of the host ligand. These results contradict the conventional description of the reaction pathway and underscore the sensitivity of rare earth solvation environment to electrolyte conditions, which is key to fundamentally understanding the electron transfer process during the rare earth reduction in supported electrolyte systems.

AWARD:

- Preproposal application for the Office of Science Early Career Researcher Program under Basic Energy Science Heavy Elements research area was encouraged for a full proposal application.



Cyclic voltammetry of 0 mM (dashed line) and 100mM neodymium chloride (solid line) in lithium borohydride/tetrahydrofuran using platinum working electrode at 20mV/s scan rate. This figure shows the irreversible reduction of neodymium metal electrodeposition in the lithium-supported electrolyte.



CONDENSED MATTER PHYSICS AND MATERIALS SCIENCE

Advanced materials with improved performance in extreme environments are critical to achieve our clean energy and security objectives. Therefore, the lab is actively working to shift the paradigm from design-build-test to digital design and manufacturing for nuclear fuels, lightweight materials and advanced survivability materials. To achieve this, we are leveraging existing expertise in metallurgy, materials and materials performance in extreme environments with innovation in advanced manufacturing and AI. Our work will enable next generation materials performance using AI coupled with advanced manufacturing to predict materials degradation ranging from the microscale to the mesoscale in a wide range of extreme service environments with process-informed physics.

CORE CAPABILITIES

Applied Mathematics

Applied Materials Science & Engineering

Computational Science

Condensed Matter Physics and Materials Science

Earth and Energy Systems and Infrastructure Analysis and Engineering

Mechanical Design and Engineering

Nuclear Engineering

AMEY KHANOLKAR

Mass transport in extreme environments—the role of the grain boundary

PROJECT NUMBER:
22A1059-067FP

TOTAL APPROVED AMOUNT:
$1,185,000 over 3 years

PRINCIPAL INVESTIGATOR:
Laura Hawkins

CO-INVESTIGATORS:
Daniel Murray, INL
Jia-Hong Ke, INL
Ruchi Gakhar, INL
Sourabh Kadambi, INL
Trishelle Copeland-Johnson, INL
Lin Shao, Texas A&M University
Lingfeng He, North Carolina State University

COLLABORATORS:
North Carolina State University
Texas A&M University

Experimental combination of applied stress, molten salt corrosion, and irradiation identified elemental mass transport in a simulated nuclear reactor environment.

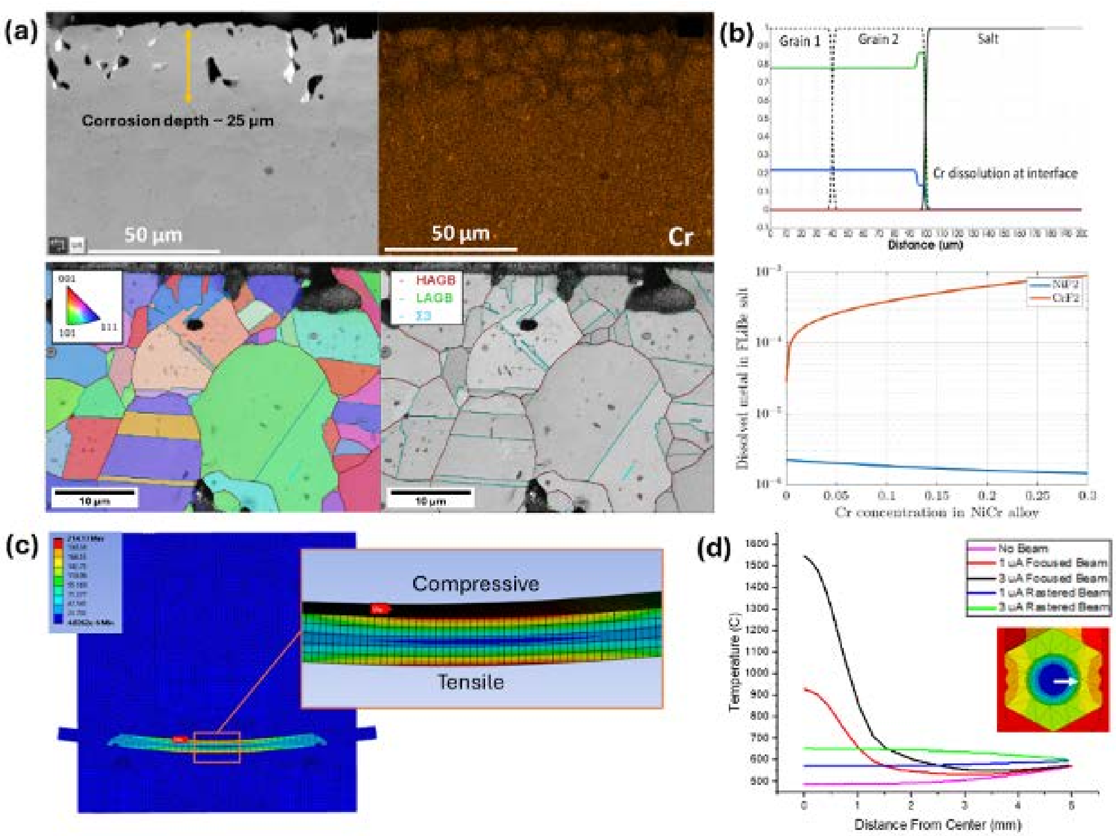
Corrosion of reactor structural materials in advanced molten salt reactors is mainly associated with the mass transport of impurities, fission products, and alloying elements along the grain boundaries, which can cause significant chemical redistribution and contribute to failure. Several alloy types were progressively subjected to extreme environments such as radiation, stress, and corrosion to study the unit mechanisms governing defect interaction at grain boundaries as a function of grain boundary character and chemistry. A new method of in situ irradiation plus corrosion was designed and optimized to mitigate temperature differentials within the corrosion chamber and increase experiment repeatability. It was proven that grain boundary character impacts a material's propensity to deplete alloying elements such chromium (Cr) with the attack of high angle grain boundaries more likely than low angle or twin grain boundaries. These results were verified with phase-field models using MOOSE for both the irradiated and corroded specimens, showing the suppression of radiation-induced segregation at these boundaries. Experimental and one-dimensional simulations of the nickel (Ni)-Cr alloys proved materials with higher percentages of Cr experience the greatest mass loss. In a dual phase system, the molten salt preferentially attacked the phase enriched in Cr, leaving the secondary phase relatively intact and mass transport across the phase boundaries low. Collaborations with Texas A&M University and North Carolina State University were established to use their ion beam laboratory and advanced materials characterization, respectively.

TALENT PIPELINE:

- Chaitanya Bhave, postdoc at INL
- Kyle Holloway, student at North Carolina State University
- Laura Hawkins, student at Texas A&M University, converted to staff
- Robert Gentile, student at North Carolina State University
- Ryan Bedell, student at Rensselaer Polytechnic Institute
- Sourabh Kadambi, postdoc at INL, converted to staff
- Trishelle Copeland-Johnson, postdoc at INL, converted to staff
- Yinyin Hong, student at Texas A&M University

PUBLICATIONS:

- M. Song, J. Yang, X. Liu, L. Hawkins, Z. Jiao, L. He, Y. Zhang, D. Schwen, X. Lou, “Void swelling in additively manufactured 316L stainless steel with hafnium composition gradient under self-ion irradiation,” J. Nucl. Mater. 578 (2023).
- J. Yang, L. Hawkins, L. He, S. Mahmood, M. Song, K. Schulze, X. Lou, “Intragranular irradiation-assisted stress corrosion cracking (IASCC) of 316L stainless steel made by laser direct energy deposition additive manufacturing: delta ferrite-dislocation channel interaction,” J. Nucl. Mater. 577 (2023).
- L. Hawkins, J. Yang, M. Song, D. Schwen, Y. Zhang, L. Shao, X. Lou, L. He, “The effect of secondary phases on microstructure and irradiation damage in an as-built additively manufactured 316L stainless steel with a hafnium compositional gradient,” J. Nucl. Mater. 587 (2023).
- J. Yang, L. Hawkins, Z. Shang, E. McDermott, B. Tsai, L. He. Y. Lu, M. Song, H. Wang, X. Lou, “Dislocation channel broadening – a new mechanism to improve irradiation-assisted stress corrosion cracking resistance of additively manufactured 316L stainless steel,” Acta Mater. 266 (2024).



(a) Effects of grain boundary character on mass transport of Cr during molten salt corrosion, (b) One-dimensional multi phase-field simulation of Ni-Cr corrosion showing Cr dissolution in the salt, (c) Ansys simulation of in situ corrosion and stress on a 4-point bend specimen and (d) temperature effects on a sample in the in situ corrosion and irradiation cell.

Local measurement of lattice anharmonicity in strongly correlated systems: toward validation of advanced electronic structure calculations

PROJECT NUMBER:
22A1059-089FP

TOTAL APPROVED AMOUNT:
$797,000 over 3 years

PRINCIPAL INVESTIGATOR:
Amey Khanolkar

CO-INVESTIGATORS:
Caleb Picklesimer, INL
Cody Dennett, INL
Robert Schley, INL
Shuxiang Zhou, INL
Marat Khafizov, The Ohio State University
Stephen Scott Parker, Los Alamos National Laboratory

COLLABORATORS:
Air Force Research Laboratory
University of Utah

The first experimental evidence of sub-nanosecond anharmonicity-induced structural phase transition in strontium titanate opens new avenues for ultrafast switching in the information processing and energy storage sectors.

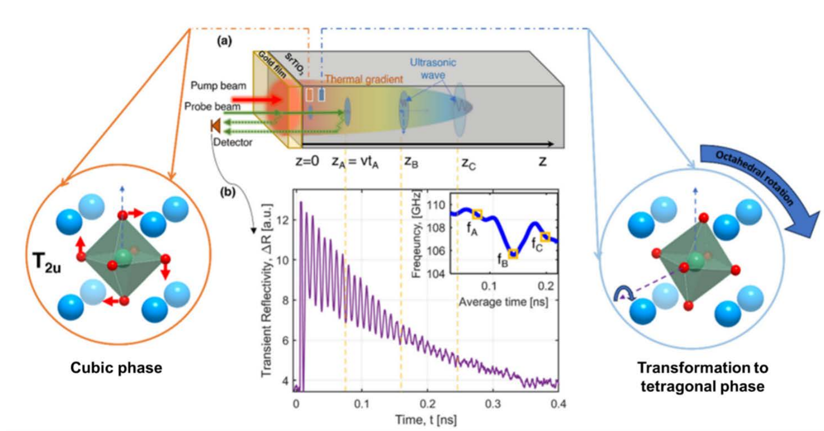
This project demonstrated that ultrafast laser excitation and pump-probe optical spectroscopy techniques are effective in monitoring the timescale of lattice anharmonicity-driven structural phase transitions in materials relevant to the energy industry. A new experimental capability for performing simultaneous low temperature (from room temperature down to 10 K) and high pressure (from atmospheric pressure up to 30 GPa) optical spectroscopy measurements was developed. This enabled the measurement of temperature- and pressure-induced changes to phonon-mediated physical properties and provided a platform for extracting the isobaric and isothermal Gruneisen parameters that represent contributions from anharmonic terms of the interatomic force. Local measurements of phonon anharmonicity were performed under two external stimuli—cryogenic temperature and applied hydrostatic pressure. In the first instance, time-domain Brillouin scattering, an ultrafast pump-probe spectroscopy technique, monitored the dynamics of the structural phase transition in strontium titanate (SrTiO3), a perovskite oxide band insulator relevant to many sectors of the energy industry. Using femtosecond optical pulses from the pump laser, a thermally-driven tetragonal-to-cubic structural transformation was initiated, and the structure of the crystal was detected through changes in the frequency of Brillouin oscillations induced by propagating acoustic phonons. By coupling the measured Brillouin oscillation frequency to a spatiotemporal heat diffusion model, it was demonstrated that deposition of sufficient thermal energy induced a rapid transformation to the cubic phase within the heat-affected region for the SrTiO3 in the low temperature tetragonal phase. The initial phase change was followed by a slower cubic-to-tetragonal reversal that occurred on a timescale of hundreds of picoseconds. Evidence of such a fast structural transition in perovskites can open new avenues in the areas of information processing and energy storage. The second instance involved using Raman spectroscopy to monitor frequency and linewidth changes of the zone-centered optical phonon in ceria induced by externally-applied hydrostatic pressure. Such measurements offer stringent tests to density functional theory and dynamical mean-field theory predictions of thermo-physical properties in complex materials and can accelerate their deployment in new energy generation technologies. A research collaboration with the University of Utah was established during this project to build expertise in high pressure experimentation at INL.

TALENT PIPELINE:

- Desta Tewabe, student at University of Southern California
- Md. Minaruzzaman, student at The Ohio State University
- Saqeeb Adnan, student at The Ohio State University

PRESENTATIONS AND PUBLICATION:

- Adnan, S., Khanolkar, A., Zhou, S., Hurley, D.H. and Khafizov, M., "Optical pulse-induced ultrafast antiferrodistortive transition in SrTiO3," Applied Physics Reviews, 11(3), 2024.
- Adnan, S., Khanolkar, A., Hurley, D.H., Khafizov, M., "Evolution of Vibrational Modes during Antiferrodistortive Phase Transition in SrTiO3," TMS 2024, 153rd Annual Meeting & Exhibition, March 3 – 7, 2024, Orlando, FL.
- Adnan, S., Khanolkar, A., Hurley, D.H., Khafizov, M., "Ultrafast optical probing of temperature-induced antiferrodistortive phase transition in SrTiO3," 2023 Materials Research Society (MRS) Fall Meeting & Exhibit, November 26 – December 1, 2023. Boston, MA.
- Hurley, D.H., Khanolkar, A., Khafizov, M., Adnan, S., "Probing low temperature phonon properties of SrTiO3 using picosecond ultrasounds," Invited talk at the 2023 International Congress on Ultrasonics, September 18 – 21, 2023, Beijing, China.
- Khanolkar, A., Adnan, S., Hurley, D.H., Khafizov, M., "Ultrafast characterization of the antiferrodistortive transition in strontium titanate," Accepted for presentation at the IEEE Nanotechnology Materials and Devices Conference (NMDC), October 22 – 24, 2024, Salt Lake City, UT.



(a) Formation of a thermal gradient due to heating of the SrTiO3 sample surface by the pump laser beam. Rapid thermal expansion of the transducer layer (gold film) launches coherent acoustic phonons that propagate along the sample depth normal to its surface across the region with the thermal gradient. The atomic configurations represent phonon vibrational modes across the sample depth. The hot cubic region has the triply degenerate T2u vibrational mode characterized by the planar rotation of oxygen octahedra, which is impacted during the cubic-to-tetragonal transition. (b) Time-resolved reflectivity change from the time-domain Brillouin scattering measurements on the StTiO3 single crystal at 92 K, that highlights the principle of depth profiling using the frequency of the generated coherent acoustic phonons.

Understanding deformation and phonon transport mechanisms in irradiated high entropy carbide ceramics

PROJECT NUMBER:
22A1059-107FP

TOTAL APPROVED AMOUNT:
$750,000 over 3 years

PRINCIPAL INVESTIGATOR:
Kaustubh Bawane

CO-INVESTIGATORS:
Linu Malakkal, INL
Zilong Hua, INL
Bai Cui, University of Nebraska–Lincoln
Lingfeng He, North Carolina State University
Yongfeng Lu, University of Nebraska–Lincoln

COLLABORATOR:
University of Tennessee–Knoxville

High entropy ceramic carbides exhibit high radiation tolerance, making them viable solutions for next generation nuclear reactors.

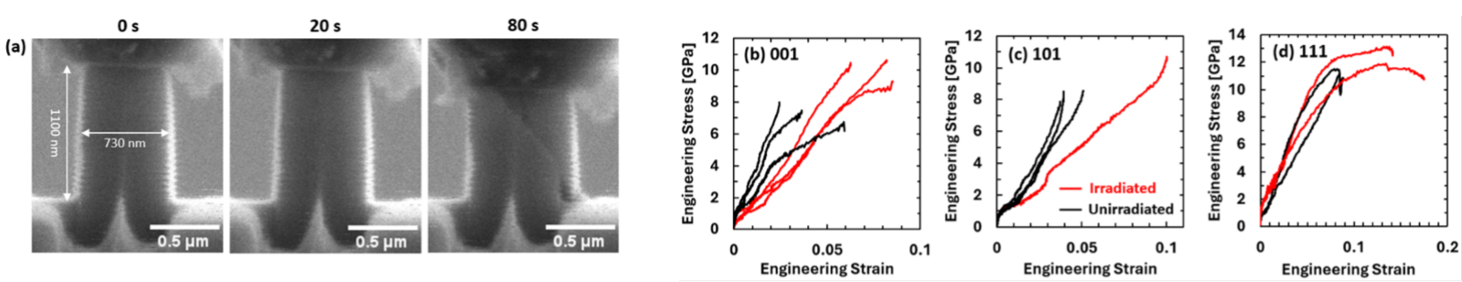
Traditional ceramic materials used in nuclear applications, such as silicon carbide and zirconium carbide, are known for their ability to withstand high temperatures better than metallic alloys like stainless steel and nickel alloys. However, these ceramics still face challenges, including limited resistance to radiation damage and less than ideal thermal and mechanical properties. To address these issues, this project used entropy stabilization to design and synthesize new ceramic materials that are more resistant to radiation and can tolerate higher temperatures. The team used entropy stabilization to create opportunities for design and synthesis of novel irradiation-resistant and high-temperature tolerant ceramic materials for nuclear applications. The high configurational entropy enhanced the simultaneous solubility of many cation elements (four or more), which were selected to optimize the physical properties such as thermal conductivity, mechanical properties, and irradiation resistance. Through the investigation of the fundamental mechanisms of deformation/fracture and phonon transport mechanisms in high entropy carbide ceramics via experiments and simulations, the team designed and discovered new ceramic materials for extreme environments. During this project, the team synthesized high entropy carbides using spark plasma and laser sintering methods and measured the thermal properties using various techniques. To further understand the material thermal properties, density functional theory simulations computed phonon dispersion spectra of high entropy carbides. Additionally, advanced characterization techniques confirmed that all samples have single phase face-centered cubic structure. The synthesized samples were then subjected to irradiation with heavy ions to stimulate radiation damage and compared with the single-element systems. The synthesized samples were irradiated with heavy ions to simulate radiation damage and compared with single-element systems. Mechanical behavior was studied using micropillar compression testing, revealing orientation-dependent variations in yield stress and critical resolved shear stress due to specific slip systems activated during deformation. The study found that irradiated pillars generally exhibit increased hardness due to the formation of irradiation-induced defect clusters, which act as barriers to dislocation motion during deformation. Overall, the irradiated high entropy carbide ceramics exhibit high radiation tolerance with minimal radiation-induced defects, suggesting that high entropy carbide ceramics could be a viable solution for next generation nuclear reactors with superior radiation and mechanical resistance.

TALENT PIPELINE:

- Bao-Phong Nguyen, student at University of Wisconsin–Madison
- Lanh Trinh, student at University of Nebraska–Lincoln
- Luke Wadle, student at University of Nebraska–Lincoln
- Samuel Ruiz, student at University of Nebraska–Lincoln
- William Reed, student at Texas A&M University

PRESENTATIONS AND PUBLICATIONS:

- Cody A. Dennett, Zilong Hua, Eric Lang, Fei Wang, Bai Cui, "Thermal conductivity reduction in (Zr0.25Ta0.25Nb0.25Ti0.25) C high entropy carbide from extrinsic lattice defects," Materials Research Letters, 10:9 (2022), 611-617.
- Lanh Trinh, Zilong Hua, Kaustubh Bawane, Lingfeng He, Linu Malakkal, Xin Chen, Luke Wadle, Yongfeng Lu, Bai Cui, "Selective laser sintering and spark plasma sintering of (Zr, Nb, Ta, Ti, W)C compositionally complex carbide ceramics," Journal of the American Ceramics Society, 1-14, 2024.
- Lanh Trinh, Fei Wang, Kaustubh Bawane, Khalid Hattar, Zilong Hua, Linu Malakkal, Lingfeng He, Luke Wadle, Yongfeng Lu, Bai Cui, "Compositionally complex carbide ceramics: A perspective on irradiation damage," Journal of Applied Physics, 2024; 135(20): 200901.
- Kaustubh Bawane, Lanh Trinh, Fei Teng, Zilong Hua, Linu Malakkal, Samuel Ruiz, Fei Wang, Bai Cui, and Lingfeng He, "Investigating the effects of irradiation on microstructure, micromechanical and thermal properties of high entropy carbide ceramics," Invited presentation at Material Science and Technology Conference (MS&T), Columbus, Ohio, 2023.
- Bai Cui, Lanh Trinh, Zilong Hua, Kaustubh Bawane, Lingfeng He, Linu Malakkal, Xin Chen, Luke Wadle, Yongfeng Lu, "Selective laser sintering and spark plasma sintering of compositionally complex carbide ceramics," Invited presentation at Material Science and Technology Conference (MS&T) Pittsburgh Pennsylvania, 2024.
- Linu Malakkal, Kaustubh Bawane, Lanh Trinh, Fei Teng, Zilong Hua, Samuel Ruiz, Fei Wang, Bai Cui, Lingfeng He, "Irradiation Effects on the Microstructure, Micro Mechanical and Thermal Properties of HECC," Invited presentation at Material Science and Technology Conference (MS&T) Pittsburgh Pennsylvania, 2024.
- Zilong Hua, Kaustubh Krishna Bawane, Linu Malakkal, Lanh Trinh, Xin Chen, Luke Wadle, Bai Cui, Lingfeng He, and Cody Dennett, "Experimental investigation of thermal conductivity of high entropy ceramics," Invited presentation at the Pan American Ceramic Congress and Ferroelectrics Meeting of Americas, Panama City, Panama, 2024.
- Linu Malakkal, Kaustubh K Bhawane, Cody A Dennet, Zilong Hua, Lingfeng He, Yongfeng Lu, and Bai Cui, "Phonon broadening and thermal conductivity high entropy ceramic carbide," Invited presentation at the Material Science and Technology Conference Pittsburgh Pennsylvania, 2022.
- Bai Cui, Kaustubh Bawane, Linu Malakkal, Zilong Hua, and Lingfeng He, "Will "high entropy" carbides be enabling materials for extreme environments?" TMS, Florida, USA, 2024.
- Bai Cu, Fei Wang, Lanh Trinh, Luke Wadle, Yongfeng Lu, Kaustubh Bawane, Zilong Hua, Linu Malakkal, Lingfeng He, Cody Dennett, and Frederic Monteverde, "Will high-entropy carbides be enabling materials for extreme environments?" Material Science and Technology Conference, Columbus, Ohio, 2023.



(a) Scanning electron micrographs showing the irradiated 001 pillar at different stages within the compression tests. (b)–(d) Engineering stress-strain curves of irradiated and unirradiated zirconium tantalum niobium titanium carbide micropillars along 001 , 101 , and 111 crystallographic orientations. The study finds that irradiated pillars generally exhibit increased hardness due to the formation of irradiation-induced defect clusters, which act as barriers to dislocation motion during deformation.

Design, growth, and characterization of actinide nitride thin films

PROJECT NUMBER:
22P1066-004FP

TOTAL APPROVED AMOUNT:
$240,000 over 3 years

PRINCIPAL INVESTIGATOR:
Kevin Vallejo

CO-INVESTIGATOR:
David Hurley, INL

COLLABORATORS:
Los Alamos National Laboratory
National Institute of Standards and Technology
Stanford Linear Accelerator
Universidad Nacional Autónoma de México
Western Michigan University

INL scientists established the first molecular beam epitaxy system equipped to grow actinide-based ceramic thin films.

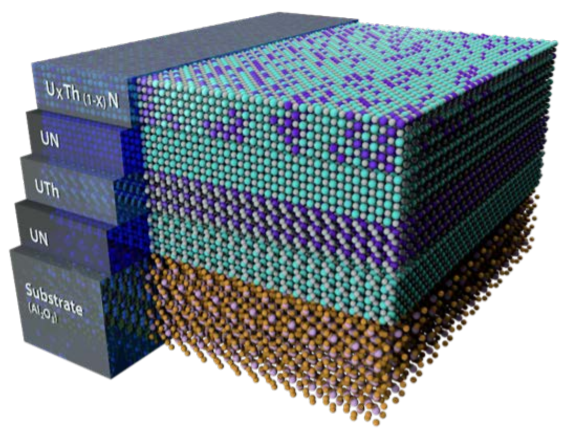
Physical properties of materials determine their selection and performance, especially when included in activities that require strict safety measures such as nuclear power reactors. Samples of materials that contain impurities and defects make the determination of their physical properties difficult, often requiring several measurements and intensive computational calculations to determine the real values of, e.g., thermal conductivity. Using molecular beam epitaxy, we established the controls, processes, and safety procedures necessary to synthesize thin films of actinide-based ceramic nuclear fuel materials. As part of the development of these controls, synthesis of transition metal- and lanthanide-based ceramics was also accomplished.

TALENT PIPELINE:

- Kevin Vallejo, Russell L. Heath distinguished postdoctoral fellow at INL, converted to staff

PUBLICATION:

- Vallejo, Kevin D., Firoza Kabir, Narayan Poudel, Chris A. Marianetti, David H. Hurley, Paul J. Simmonds, Cody A. Dennett, and Krzysztof Gofryk, "Advances in actinide thin films: synthesis, properties, and future directions," Reports on Progress in Physics 85, no. 12 (2022): 123101.



Sketch of a proposed heterostructure consisting of atomically precise uranium nitride (UN), uranium thorium (UTh), and uranium thorium nitride (UxTh(1-x)N) thin films on an aluminum oxide (Al2O3) substrate.

Electronic and thermodynamic properties of unconventional superconductors

PROJECT NUMBER:
22P1066-009FP

TOTAL APPROVED AMOUNT:
$300,000 over 2 years

PRINCIPAL INVESTIGATOR:
Krzysztof Gofryk

CO-INVESTIGATOR:
Sabin Regmi, INL

COLLABORATORS:
Lawrence Berkeley National Laboratory
University of Central Florida

INL researchers measure the electronic structure of novel 4f and 5f-electron topological quantum materials for the first time.

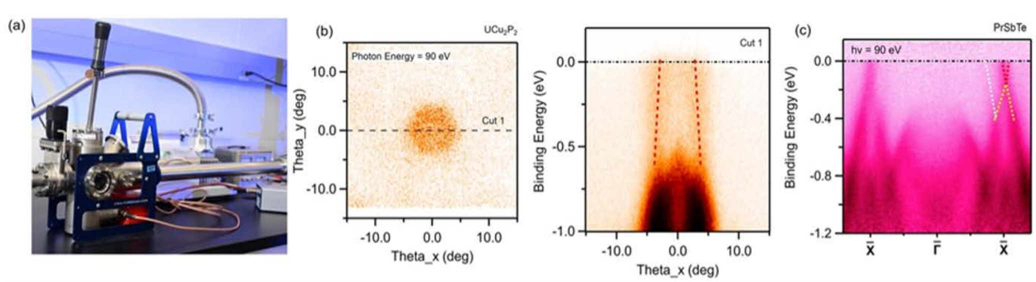
This project led to the successful development of Ferrovac ultra-high vacuum suitcase to work with the angle-resolved photoemission spectroscopy setups at the University of Central Florida and Advanced Light Source synchrotron at Lawrence Berkeley National Laboratory. To work with radioactive elements at these facilities, the team developed all necessary documentation and safety procedures to study low temperature magnetic, transport, and thermodynamic properties of selected 4f and 5f-electron quantum materials focusing on uranium compounds (U6X) where X = manganese (Mn), cobalt (Co), and iron (Fe)], uranium copper phosphate (UCu2P2), and praseodymium antimony telluride (PrSbTe). Through successful use of the vacuum suitcase, the team completed angle-resolved photoemission spectroscopy measurements on single crystals of 5f-electron system UCu2P2 at the Advanced Light Source and were able to obtain first-of-kind measurements of this material's Fermi surface and underlying electronic structure. The team also used this technique to study the 4f-electron based topological material PrSbTe and successfully revealed the presence of topological nodal-line states. These results open exciting avenues to extend the studies to uranium-based systems and actinide systems in general to look for topological states that have not been explored and their potential applications.

TALENT PIPELINE:

- Sabin Regmi, postdoc at INL

PUBLICATIONS:

- S. Regmi, I. B. Elius, A. P. Sakhya, M. Sprague, M. I. Mondal, V. Buturlim, K. Booth, T. Romanova, K. Gofryk, A. Ptok, D. Kaczorowski, and M. Neupane, "Electronic structure in a rare-earth based nodal-line semimetal candidate PrSbTe," Physical Review Materials 8, L041201 (2024).
- K. D. Vallejo, F. Kabir, N. Poudel, C. A. Marianetti, D. H. Hurley, P. J. Simmonds, C. A. Dannett, and K. Gofryk, "Advances in actinide thin films: synthesis, properties, and future directions," Reports on Progress in Physics 85, 123101 (2022).



(a) Personalized Ferrovac ultra-high vacuum suitcase. (b) Fermi surface of uranium-based pnictide system UCu2P2 (left) and energy-momentum band dispersion along cut 1 (right). (c) Angle-resolved photoemission spectroscopy result on band structure of rare earth based nodal-line semimetal PrSbTe.

Understanding the role of fission products and complex defect structures in the gas bubble superlattice formation and collapse mechanisms

PROJECT NUMBER:
22P1066-010FP

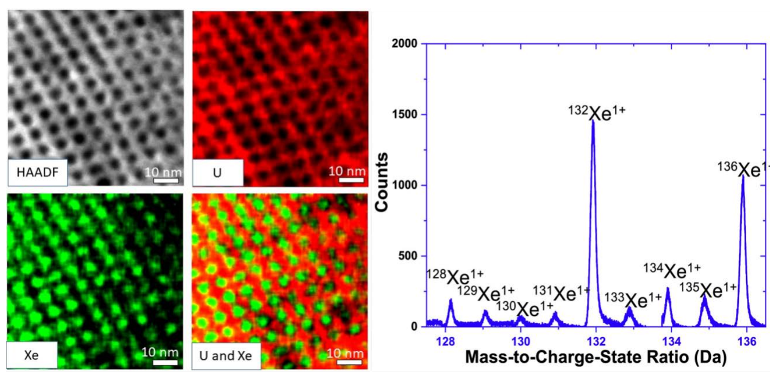
TOTAL APPROVED AMOUNT:
$360,000 over 3 years

PRINCIPAL INVESTIGATOR:
Mukesh Bachhav

CO-INVESTIGATORS:
Brandon Miller, INL
Dennis Keiser, INL
Sohail Shah, INL
Charlyne Smith, University of Florida

Gas bubble superlattices in metallic nuclear fuel confine xenon.

One of the key areas in the development of low enriched uranium metallic fuels is the evolution and behavior of fission gases. Fission gas bubble superlattices in metallic fuels have been postulated to consist of xenon (Xe) atoms, although its chemical and physical arrangement within the gas bubble superlattice has not been verified. In this project, Xe was chemically profiled using atomic resolution atom probe tomography and scanning transmission electron microscopy. These complementary techniques provided conclusive evidence of Xe presence in irradiated uranium-molybdenum (U-Mo) fuels. A correlative analysis approach using transmission electron microscopy and atom probe tomography was used in this study. A quantitative assessment of Xe content within gas bubble superlattices found fission densities up to 4.5 × 1021. The chemical composition of Xe within the bubble superlattice was found to be up to 8 ± 1.3 atomic % (at. %). These scanning transmission electron microscopy results were complimented by atom probe tomography data showing Xe distribution within the irradiated U-Mo fuel sample. This project highlighted the evidence of Xe within atomically arranged bubble superlattice and provided fundamental insights into the physical state of Xe in low enriched U-Mo monolithic fuel.



Left: High resolution studies using scanning transmission electron microscopy and atom probe tomography. Right: Time of flight mass spectrometry performed on irradiated U-Mo specimen to confirm presence and distribution of Xe.

TALENT PIPELINE:

- Sohail Shah, postdoc at INL

PRESENTATIONS AND PUBLICATIONS:

- C. Smith, K. Bawane, J. Gan, D. Keiser, D. Salvato, M. Bachhav, J.-F. Jue, "The role of UC inclusions in the development of fission gas bubble superlattice neutron-irradiated monolithic U-10Mo fuels," Journal of Nuclear Materials, 581, 154474.
- C. Smith, J.-F. Jue, T. Trowbridge, D. Keiser, J. Madden, A. Robinson, J. Giglio, "Possible impacts of Mo chemical banding and second phase impurities on the irradiation behavior of monolithic U-10Mo fuel," Journal of Nuclear Materials, 576, 154264.
- B. Miller, M. Bachhav, B. Kombaiah, C. Smith, A. Aitkaliyeva, L. He, D. Keiser, J. Madden, A. Robinson, J. Gan, "Evidence of Xe-incorporation in the bubble superlattice in irradiated U-Mo fuel," Journal of Nuclear Materials, 587, 154743.
- D. Salvato, C. Smith, T. Pavlov, W. Hanson, K. Bawane, M. Bachhav, B. Miller, T. Trowbridge, J. Gan, J. Giglio, A. Winston, J. Henley, A. Robinson, D. Keiser, I. Glagolenko, B. Ye, Z.-G. Mei, L. Jamison, G. Hofman, A. Yacout, S. Van den Berghe, A. Leenaers, "Contributions to the mechanistic understanding of the microstructural evolution in irradiated U-Mo dispersion fuel," Journal of Nuclear Materials, 588, 154789.
- S. Shah, D. Salvato, C. Smith, T. Yao, F. Teng, J. Giglio, M. Bachhav, "Advanced Characterization of Metallic fuels by Atom Probe Tomography," Microscopy & Microanalysis Conference, July 28 – August 1, 2024.
- A. Pradhan, S. Shah, T. Yao, I. Charit, M. Bachhav, "Atom Probe Tomography (APT) characterization of Annular U-Zr metallic fuel cladded with HT-9," Microscopy & Microanalysis Conference, July 28 – August 1.
- M. Bachhav, A. Kamboj, S. Shah, J. Gan, J. Giglio, F. Cappia, F. Teng, "Precision burnup assessment through dynamic peak fitting in atom probe tomography for metallic and ceramic fuel analysis," Microscopy & Microanalysis Conference, July 28 – August 1.

Synthesis of undiscovered ternary nitride compounds via molecular beam epitaxy

PROJECT NUMBER:
22P1067-001FP

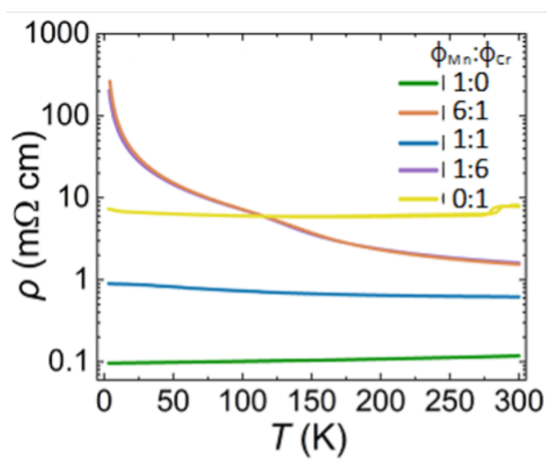
TOTAL APPROVED AMOUNT:
$455,000 over 3 years

PRINCIPAL INVESTIGATOR:
Brelon May

COLLABORATORS:
Boise State University
National Institute of Standards and Technology
SLAC National Accelerator Laboratory
University of Hawaii
Universidad Nacional Autónoma de México
Western Michigan University

Molecular beam epitaxy synthesis of novel ternary nitride thin films demonstrates significant progress in materials development and fosters a strong foundation for future funding and research.

This project was the primary support for a strategic hire to develop a research program around the new molecular beam epitaxy tool installed at INL. The goals of this project were to synthesize ternary nitrides via molecular beam epitaxy and provide a platform for establishing INL as a location for the fabrication of high quality single crystalline thin films. Significant progress was made in the examination of several transition metal nitride systems via molecular beam epitaxy such as the binary manganese, chromium, nickel, zirconium, tantalum, and gallium nitrides (MnN, CrN, NiN, ZrN, TaN, and GaN) on various substrates. After proper processing parameters for single phase samples were elucidated, investigations were extended to select ternaries such as manganese chromium nitride (MnCrN), manganese nickel nitride (MnNiN). Various phases of MnxNy were synthesized epitaxially. The physical properties of CrN thin films were found to be strongly dependent on the deposition temperature, and the reported magnetostructural transition ~280 K was observed in these films. The previously unreported ternary MnxCr1-xN system was synthesized for the first time and showed a variation of resistivity covering over ten orders of magnitude at low temperatures. Superconducting ZrN thin films were also deposited by molecular beam epitaxy for the first time. The critical points showed a substantial dependence on the growth temperature. This work also supported the development of some select rare earth binary nitrides. This project involved five graduate students on-site and another who remained at their university and an INL postdoc who was converted to staff. This work provided the foundation for the pursuit of more than 20 internal and external funding opportunities.



Electrical resistivity as a function of temperature for manganese (Mn) chromium (Cr) nitride alloys with different supplied flux ratios (ϕMn:ϕCr) during deposition.

TALENT PIPELINE:

- Ahmed Wasif Mustakim, student at University of California Davis
- Jennyfer Vivas Gomez, student at University of Central Florida
- Kevin Vallejo, Russell L. Heath distinguished postdoctoral fellow at INL, converted to staff
- Nicole Fessler, student at Cornell
- Trent Garrett, student at Boise State University
- Zach Cresswell, student at University of Minnesota

PRESENTATIONS AND PUBLICATIONS:

- Kevin D. Vallejo, Zachery Cresswell, Volodymyr B Buturlim, Brian Newell, Krzysztof Gofryk, Brelon J. May, "Synthesis of samarium nitride thin films on magnesium oxide (001) substrates using molecular beam epitaxy," Crystals, 2024, 14(9), 765.
- Brelon J. May, Sabin Regmi, Amey R Khanolkar, Volodymir B Buturlim, Zachery Cresswell, Kevin Vallejo, Krzysztof Gofryk, David H Hurley, "Molecular Beam Epitaxy of Superconducting Zirconium Nitride on GaN Substrates," AIP Advances (under review).
- Brelon J. May, "A Route Towards Actinide Heterostructure Synthesis and Science," North American MBE (NAMBE), September 2022.
- Brelon J. May, "Molecular Beam Epitaxy of Novel Nitrides," MRS, November 2022.
- Brelon J. May, "A Path Toward Molecular Beam Epitaxy of Single Crystalline Actinide Materials," APS, March 2023.
- Brelon J. May, "Novel Nitrides as a path toward synthesis of actinide thin films," American Conference on Crystal Growth and Epitaxy (ACCGE), August 2023.
- Brelon J. May, "Molecular beam epitaxy of binary and ternary manganese and chromium nitrides," NAMBE, September 2023.
- Kevin D. Vallejo, "Transport of rare earth nitrides deposited via molecular beam epitaxy, NAMBE, September 2023.
- Kevin D. Vallejo, "Integration of epitaxial ZrN with GaN: superconductors and wide bandgaps," NAMBE, September 2023.
- Brelon J. May, "Molecular beam epitaxy of cubic nitrides," Invited presentation at TMS, March 2024.
- Kevin D. Vallejo, "Manganese chromium nitride thin film synthesis via molecular beam epitaxy" APS, March 2024.
- Kevin D. Vallejo, "Thin film synthesis of rare earth and actinide nitrides using MBE," ACCGE, June 2024.
- Brelon J. May, "Epitaxial synthesis and integration of cubic nitrides," Invited presentation at Pacific Northwest chapter of AVS (PNWAVS), August 2024.

Understanding defect dynamics in complex structural alloys

PROJECT NUMBER:
23P1078-001FP

TOTAL APPROVED AMOUNT:
$603,634 over 2 years

PRINCIPAL INVESTIGATOR:
Yanwen Zhang

CO-INVESTIGATOR:
Boopathy Kombaiah, INL

COLLABORATORS:
Guangdong University of Technology, China
Los Alamos National Laboratory
Lukasiewicz Research Network – Institute of Microelectronics and Photonics, Poland
National Centre for Nuclear Research, Poland
Nelson Mandela University, South Africa
Oak Ridge National Laboratory
Shaoxing University, China
The University of Manchester, United Kingdom
University of Tennessee, Knoxville
VTT Technical Research Centre of Finland, Finland
Warsaw University of Technology, Poland
Zhejiang University, China

Materials design based on electronic and atomic level information enables first-principles design of novel, high performance alloys.

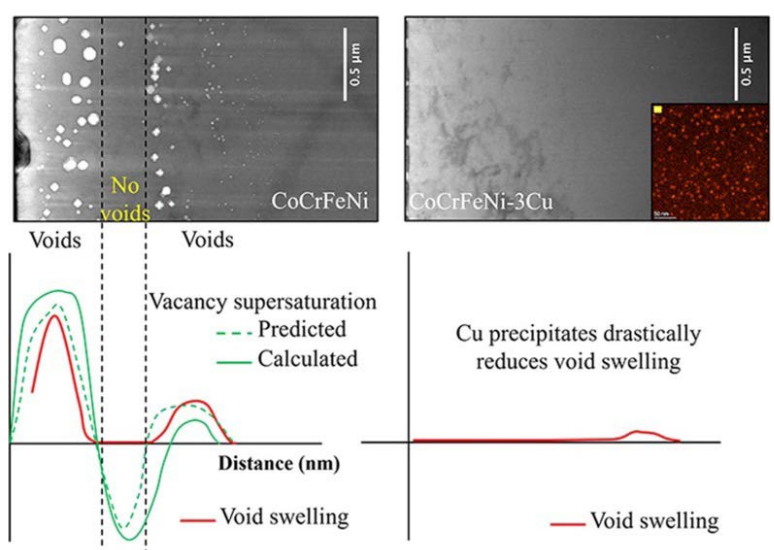
Success in synthesis of multicomponent concentrated solid-solution alloys with elements at high or near equiatomic ratios has vastly expanded the compositional space for new alloy discovery. These concentrated solid-solution alloys, including medium-entropy alloys and high entropy alloys, form nearly random solid solutions on simple underlying crystalline lattices, such as face-centered cubic and body-centered cubic. In this project, we studied the role of chemical disorder rising from multiple transition metals and effects of nanoscale inhomogeneity—nanostructured concentrated solid-solution alloys and nanolaminate face-centered cubic/body-centered cubic phases. In contrast to traditional trial-and-error approaches, our research focused on the levels of electrons and atoms, which offers a powerful avenue to investigate their chemical and physical properties. Our hypothesis was that both fundamental characteristics (valence electron count, site displacement, and atomic mass) and nanoscale inhomogeneity in concentrated solid-solution alloys can be utilized synergistically to achieve desirable radiation performance and mechanical strength. Our objective was to understand alloying effects on defect dynamics at the level of electrons, atoms, and nanoscales to control structural stability in extreme environments. With this materials design concept in mind, we developed and characterized refractory high entropy alloys with excellent void swelling and hardening resistance under irradiation as a replacement for tungsten (W). Furthermore, we expanded our understanding of the radiation effects and mechanical behavior of high entropy alloys with low concentrations of solutes. Copper (Cu) added high entropy alloys exhibit excellent swelling resistance, better than oxide dispersion steels for advanced reactor applications. We detailed the design of high entropy ceramic materials for advanced reactor applications based their physical properties. By exploiting equilibrium and non-equilibrium defect processes in concentrated solid-solution alloys, this project avoided empirical approaches and advanced our understanding of energy dissipation pathways, deformation tolerance. and phase stability in both conventional and novel alloys.

TALENT PIPELINE:

- Lia Amalia, student at University of Tennessee, Knoxville
- Sriswaroop Dasari, postdoc at INL
- Walker L Boldman, postdoc at University of Tennessee, Knoxville

PRESENTATIONS AND PUBLICATIONS:

- D. Kalita, I. Jóźwik, Ł. Kurpaska, Y. Zhang, K. Mulewska, W. Chrominski, J. O'connell, Y. Ge, W.L. Boldman, P.D. Rack, Y. Wang, W.J. Weber, and J. Jagielski, "The microstructure and He+ ion irradiation behavior of novel low-activation W-Ta-Cr-V refractory high entropy alloy for nuclear applications," Nuclear Materials and Energy 37, 101513 (2023).
- L. Amalia, Y. Li, H. Bei, Y. Chen, D. Yu, K. An, Z. Lyu, P.K. Liaw, Y. Zhang, Q. Ding, and Y. Gao, "Copper effects on the microstructures and deformation mechanisms of CoCrFeNi high entropy alloys," Applied Physics Letters 124, 141901 (2024).
- S. Dasari, B. Kombaiah, J.D. Poplawsky, M. Bachhav, P.D. Edmondson, H. Bei, R.R. Kancharla, and Y. Zhang, "Microstructural evolution in doped high entropy alloys NiCoFeCr-3X (X=Pd/Al/Cu) under irradiation," Journal of Nuclear Materials 598, 155194 (2024).
- Y. Zhang, A.R. Khanolkar, K.K. Bawane, C.A. Dennett, Z. Hua, K. Gofryk, B. Kombaiah, W. Guo, Y. Liu, and W.J. Weber, "Physical properties and their influence on irradiation damage in metal diborides and in high-entropy materials," JOM 76, 2602 (2024).
- Y. Zhang, C. Silva, T.G. Lach, M.A. Tunes, P.D. Rack, S.E. Donnelly, W.J. Weber, "Role of Electronic Energy Loss on Interface Stability of Nanostructured High-Entropy Alloys," TMS 2023 Annual Meeting & Exhibition, March 19-23, 2023, San Diego, California.
- Y. Zhang, L. Wang, W.J. Weber, "Charged Particles: Unique Tools to Study Irradiation Resistance of High-entropy Alloys," TMS 2023 Annual Meeting & Exhibition, 2023, San Diego, California.
- B. Kombaiah, S. Dasari, Y. Zhou, K. Jin, A. Manzoor, J.D. Poplawsky, J.A. Aguiar, H. Bei, D.S. Aidhy, P.D. Edmondson, and Y. Zhang, "Nanoprecipitates to Enhance Radiation Tolerance in High-Entropy Alloys," TMS 2024 Annual Meeting & Exhibition, 2024, Orlando, Florida.



Alloy design for extreme environments: Addition of Cu in small amounts to cobalt/chromium/iron/nickel (CoCrFeNi) high entropy alloy drastically lowers void swelling under irradiation.

Ultrafast charge carrier-phonon interaction and relaxation dynamics in transition metal nitrides under pressure

PROJECT NUMBER:
24A1083-011FP

TOTAL APPROVED AMOUNT:
$250,000 over 1 year

PRINCIPAL INVESTIGATOR:
Amey Khanolkar

MENTORS:
Yanwen Zhang
Dave Hurley

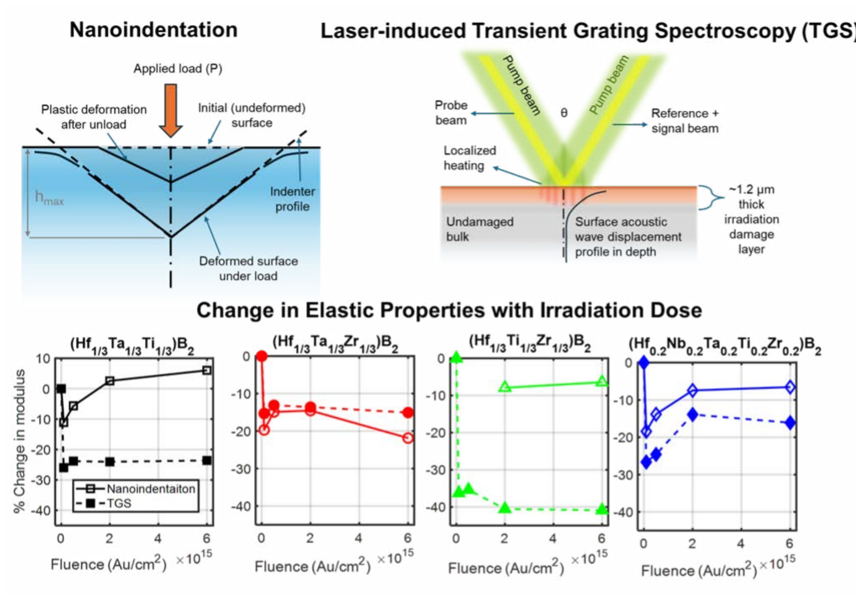
COLLABORATORS:
Yale University
Queen's University, Canada

Chemical complexity can be tailored to achieve desired carrier transport and radiation resistance in transition metal nitride and diboride ceramic material systems

Electron-phonon coupling is a fundamental interaction of quasiparticles in crystalline solids that governs their optical, electrical, transport, and thermodynamic properties. Electron-phonon coupling is known to introduce anharmonicity in the lattice potential, effectively modifying the crystal's elastic modulus. Phonon-mediated attraction between mobile electrons can also give rise to exotic phenomena such as high-temperature superconductivity. This project investigated mechanisms to tune electron-phonon coupling in transition metal nitrides and diborides, a class of compounds that show promise as radiation-tolerant structural components and as high-temperature superconductors. Intrinsic and extrinsic factors that affect electron-phonon coupling such as bond characteristics, number of valence electrons, stoichiometry, and lattice defects were studied as knobs for tuning the strength of electron-phonon coupling in crystalline or epitaxially grown transition metal nitrides and diborides. The first part of this project investigated the impact of chemical complexity in transition metal diborides on changes to two phonon-mediated physical properties—hardness and elastic modulus—caused by irradiation-induced defects. A high entropy transition metal diboride showed the greatest recovery in hardness and elastic modulus under irradiation with energetic ions to high fluences after an initial reduction in hardness and stiffness at low fluence when compared to its quaternary transition metal diboride counterparts. This highlighted the potential of tailoring defect dynamics in transition metal ceramics through lattice distortions introduced by cation size variations and chemical complexity. The second part of this project investigated charge carrier dynamics in a series of epitaxially grown zirconium nitride thin films with varying growth temperature. Transient reflectivity signals acquired at sub-nanosecond timescales following excitation of the zirconium nitride films with sub-picosecond optical pulses revealed a trend in the temporal evolution of the carrier photoexcitation, relaxation, and subsequent lattice thermalization. This result showed that growth temperature can be an effective knob for tuning electron-phonon coupling and associated thermo-physical properties of epitaxially grown transition metal nitride thin films. Results from this project highlight the potential of using chemical complexity, bond type, and synthesis conditions achieve desired carrier transport and radiation resistance in transition metal nitride and diboride ceramic material systems.

PUBLICATION:

- Khanolkar, A., Datye, A., Zhang, Y., Dennett, C.A., Guo, W., Liu, Y., Weber, W.J., Lin, H.-T., Zhang, Y., "Effects of irradiation damage on the hardness and elastic properties of quaternary and high entropy transition metal diborides," J. Appl. Phys. 136, 10, Special Issue: Era of Entropy: Synthesis, Structure, Properties, and Applications of High-Entropy Materials, 105106, 2024.



Schematics of the complimentary experimental techniques (nanoindentation and laser-induced transient grating spectroscopy) used to probe changes in the transition metal diboride elastic properties caused by irradiation-induced defects. The plots in the bottom row illustrate the percent change from an unirradiated baseline in the elastic properties of three quaternary transition metal diborides and one high entropy senary transition metal diboride. The high entropy diboride shows considerable recovery of the elastic modulus at high radiation dose, demonstrating its radiation resistance performance.

Breaking symmetry to induce unconventional superconductivity

PROJECT NUMBER:
24A1083-008FP

TOTAL APPROVED AMOUNT:
$250,000 over 1 year

PRINCIPAL INVESTIGATOR:
Brelon May

COLLABORATOR:
City College of New York

Superconducting critical points for zirconium nitride and niobium nitride thin films vary depending on deposition temperature and substrate.

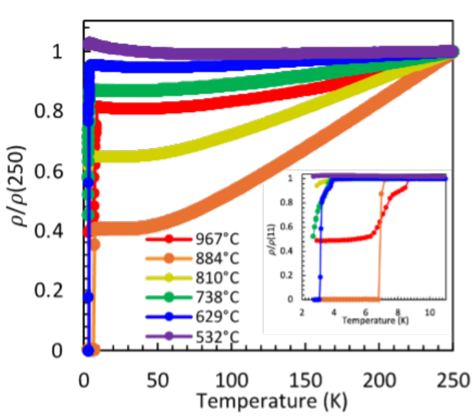
This project resulted in the successful growth of superconducting zirconium nitride and niobium nitride. The superconducting critical points of each were found to vary substantially with growth parameters such as deposition temperature and the substrate used. Maximum critical temperatures (Tc) of >8K was observed for a zirconium nitride thin film deposited at 967°C, but this sample exhibited several different Tc values and never dropped fully to zero resistance. Conversely, a deposition temperature of 884°C had a Tc of ~7K, and did drop fully to zero. Thin films of niobium nitride showed trends indicative of a high degree of electron scattering, but the best critical temperatures were >10K, and critical fields higher than 8 T. These epitaxial superconductors were then integrated with magnetic materials to investigate asymmetries in the superconducting gap. Additionally, ultrathin superlattices of superconducting and dielectric materials were deposited to investigate the thickness dependence of superconductivity. Preliminary investigations into the growth of the superconductors and antiferromagnetic chromium nitride on (100) and (111) substrates also provide a platform for integration of these materials along different crystalline directions.

TALENT PIPELINE:

- Candice Forrester, student at City College of New York
- Zach Cresswell, student at University of Minnesota

PRESENTATIONS AND INVITED TALKS

- Brelon J. May, "Molecular beam epitaxy of cubic nitrides," Invited presentation at TMS, March 2024.
- Brelon J. May, "Superconducting (001) and (111) Metal Nitrides on GaN and SiC," North American Molecular Beam Epitaxy (NAMBE), July 2024.
- Brelon J. May (invited), "Epitaxial synthesis and integration of cubic nitrides," PNWAVS, August 2024.
- Brelon J. May, "Epitaxial integration of transition metal nitrides with cubic gallium nitride," International Conference on Molecular Beam Epitaxy (ICMBE), September 2024.



Normalized resistivity of zirconium nitride thin films as a function of deposition temperature. Inset displays low temperature data to show the superconducting transitions.

"The reward of the young scientist is the emotional thrill of being the first person in the history of the world to see something or to understand something."

— Cecilia Payne-Gaposchkin, Astronomer and Astrophysicist

24-50960