INL/RPT-23-71682-Revision-0

2022 Annual Report Laboratory Directed Research & Development

March 2023

Changing the World's Energy Future

Tony Huff, Stephannie A Lambert

Idaho National Laboratory

INL is a U.S. Department of Energy National Laboratory operated by Battelle Energy Alliance, LLC

INL/RPT-23-71682-Revision-0

2022 Annual Report Laboratory Directed Research & Development

Tony Huff, Stephannie A Lambert

March 2023

Idaho National Laboratory
Idaho Falls, Idaho 83415

http://www.inl.gov

Prepared for the
U.S. Department of Energy
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517

INL/RPT-23-71682

2022

ANNUAL REPORT

Laboratory Directed Research & Development

Idaho National Laboratory

FROM INL'S CHIEF RESEARCH OFFICER

Idaho National Laboratory's (INL's) mission is "to discover, demonstrate and secure innovative nuclear energy solutions, other clean energy options and critical infrastructure." INL executes this mission through research and development across the continuum from basic science to applied science to engineering demonstration and then deployment. The Department of Energy (DOE) Laboratory Directed Research and Development (LDRD) program enables INL to conduct high-risk, impactful research that enriches the laboratory capabilities in order to further its missions. INL's LDRD portfolio specifically advances the core capabilities of the laboratory aligned with its five science and technology initiatives: 1) nuclear reactor sustainment and expanded deployment, 2) integrated fuel cycle solutions, 3) integrated energy systems, 4) advanced design and manufacturing for extreme environments, and 5) secure and resilient cyber-physical systems. The 45 projects that ended in fiscal year 2022 and highlighted in this report are just a small sample of the impressive breadth and depth of cutting-edge science, technology, and engineering ongoing at INL.

INL'S LDRD PROGRAM HAS BEEN GROWING OVER THE PAST

5 YEARS

INL's LDRD program has been growing rapidly over the past five years to advance our scientific and technical expertise related to key mission areas. In the fiscal year 2022 Annual Call for Proposals, INL continued investments in basic science related to our emerging capabilities in Chemical and Molecular Science and Condensed Matter Physics and Materials Science. Additionally, several seed LDRD projects highlight those core capabilities. As you read through this report, watch for the Chemical and Molecular Science and Condensed Matter Physics and Materials Science icons to get a sense of the research conducted at INL in these areas.

For example, LDRD-funded research in Chemical and Molecular Science advanced molten salt reactor development by adding to the fundamental understanding of salt chemistry, thereby enabling future corrosion resistant materials development.

DR. MARIANNE WALCK
Deputy Laboratory Director for Science and Technology and Chief Research Officer, Idaho National Laboratory

INL's capability and investment in Chemical and Molecular Science is key to increasing the value of nuclear energy solutions through integrated energy systems. A research project highlighted in this report discovered that gamma radiation-induced conversion of carbon dioxide and methane to carbon monoxide and hydrogen is a cost-effective, negative emissions process technology.

INL's world-class staff and unique facilities enable INL to direct research in Condensed Matter Physics and Materials Science—experimentation as well as modeling and simulation. A project in this report that studied the irradiation-induced microstructure and property changes of alpha-uranium is a great example of the power of combining experimentation with computational science. Fundamental understanding of these mechanisms advances both metallic nuclear fuels research and development and basic energy science.

Investing in secure and resilient cyber-physical systems is fundamental to INL's vision "to change the world's energy future and secure our nation's critical infrastructure." This report shares a sample of INL's deep technical expertise in analyzing cyber-physical systems. One such project used machine learning to automatically identify critical infrastructure facilities, while another used machine learning to detect malicious traffic on encrypted fifth-generation wireless networks. Other projects developed capabilities to improve critical control system security.

INL will continue to grow its LDRD program to deliver the research, development, and demonstration our nation needs to address its energy and security challenges now and in the foreseeable future. I invite you to review this report to see how INL's LDRD portfolio enables innovation, builds researcher talent, and advances INL's vision.

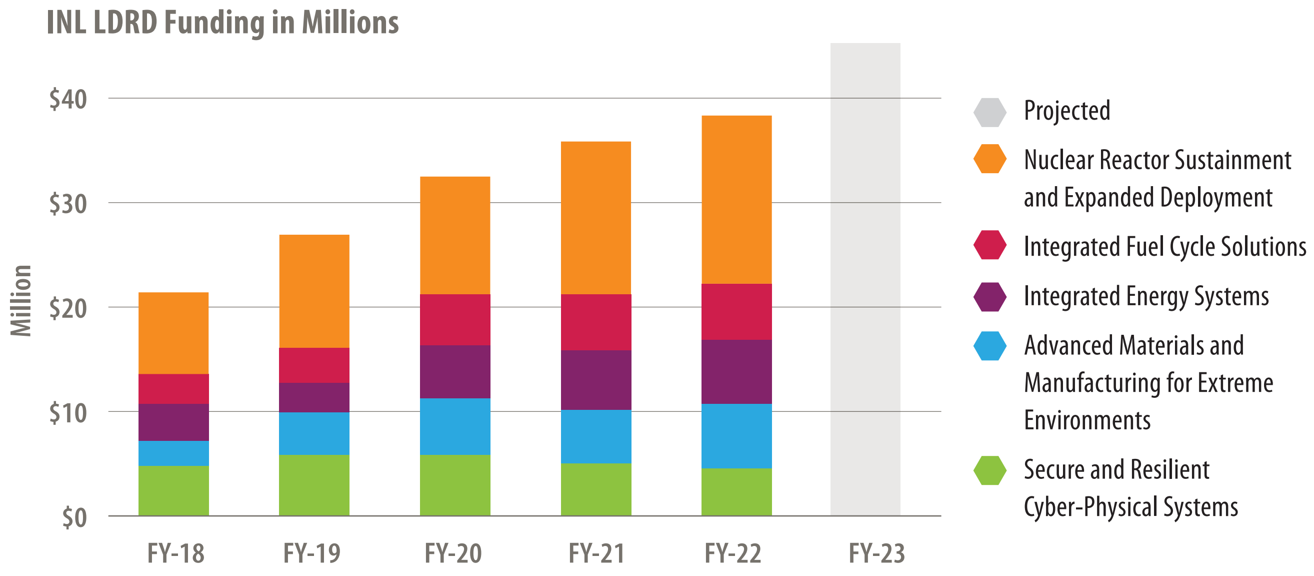

INL LDRD Funding in Millions
$40
$30
$20
$10
$0
Million
FY-18
FY-19
FY-20
FY-21
FY-22
FY-23
Projected
Nuclear Reactor Sustainment and Expanded Deployment
Integrated Fuel Cycle Solutions
Integrated Energy Systems
Advanced Materials and Manufacturing for Extreme Environments
Secure and Resilient Cyber-Physical Systems

TABLE OF CONTENTS

LDRD Overview

LDRD Overview 7

FY 2022 Program Statistics 11

Acronyms & Abbreviations 12

Nuclear Reactor Sustainment and Expanded Deployment

Beyond Nuclear Energy Materials: Utilizing Strong Spin-orbit Coupling and Topology in Actinides 16

Demonstrate Viability of Accelerated Fuel Qualification Approaches 18

Development of Novel Radiometal Chelators for Imaging and Therapeutic Nuclear Medicine 19

Combinatorial and High Throughput Materials Synthesis to Advance Nuclear Materials Discovery 20

Real-time Axial Neutron Flux Profile Measurement at The Advanced Test Reactor Critical Facility 22

Modeling and Characterization of Alpha-uranium to Accelerate Metallic Fuels Development 24

Development of a Streamlined Approach for Burnup and Microstructural Analysis of Nuclear Materials from Nanoscale to Mesoscale 26

Develop High Fidelity Computation Models to Calculate the Effective Material Properties of Porous Cells 30

Real-time Non-equilibrium Studies for Nuclear Materials Performance 33

Flash Neutron Radiography at the Transient Reactor Test Facility 34

Modeling Internal Material Melting 36

Developing Generalized Methods for Handling Transmutation Radiation Damage 37

Multi-principal Element Alloy Fuels for Fast Reactors 38

Accelerate Multiphysics Solves Using Deep Learning 40

Coupled Multiphysics Safety Analysis of Molten Salt Reactors Subjected to Earthquakes 41

Oxidation of Cerium Nitride under Irradiation 43

Development and Demonstration of Spatial Correlation Techniques among Diverse Data Sets 44

Variationally Consistent Contact Dynamics for General Nuclear Fuel Modeling 46

Understanding Non-equilibrium, Nanoscale Defects in Bulk Metals 48

Analysis of Mars Perseverance Multi-mission Radioisotopic Thermoelectric Generator Neutron/Gamma Spectrum for use on Dragonfly and Nuclear Thermal Propulsion Materials Modeling 50

Integrated Fuel Cycle Solutions

Advanced Computational Modeling and Experimental Determination of the Thermophysical Properties of Molten Salt Systems Applicable to Molten Salt Reactor Design 54

Understanding Salt Chemistry and Corrosion Control of Chloride Salts for Molten Salt Reactors 56

Development of a Simple Single Step Ultra Chemical Purification of Neptunium 58

Moving Molten Zone Refining Process Development for Innovative Fuel Cycle Solutions 59

A Novel Head-end Process for Used Uranium-aluminum Fuels 60

Integrated Energy Systems

Fundamentals for the Valorization of Carbon Dioxide Toward Integration to Nuclear Power Plants 64

Improving the Stability and Durability of Aptamer Based Smart Materials Using Extremophiles 66

Natural Gas to Chemical Intermediate by Single Step Synthesis Using Tubular Ceramic Catalytic Membrane Reactor 70

Up-cycling Process Feasibility for Coupled Radiolytic and Biochemical Conversion of Polyethylene 72

Flexible Chemical Manufacturing for Carbon Dioxide Hydrogenation to Methanol 74

Advanced Materials and Manufacturing for Extreme Environments

Multi-role High Entropy Alloys for Extreme Environments 78

Direct Integrated Advanced Manufacturing of Multi-component Solid Fuel Systems with Embedded Sensors 79

Advanced Manufacturing of Heat Pipes with Connectivity to Thermoelectrics 80

Development of a Laser Ultrasonics-based Approach for Rapid Screening of High Entropy Alloys 84

Feasibility Study of an Optical Fiber Instrument to Monitor Microstructure of Spark Plasma Sintering Products 86

Recovering Cobalt from Spent Lithium-ion Batteries Using Electrochemical Membrane Reactor without Chemical Consumption 87

Development of Scalable Design Optimization Parameters for Bi-component Protective Systems 88

Water Soluble Support Material for Directed Energy Deposition Additive Manufacturing 89

Secure and Resilient Cyber-Physical Systems

Resilient Attack Interceptor for Intelligent Devices 92

Protocol Analytics to Enable Forensics of Industrial Control Systems 94

Automated Infrastructure and Dependency Detection via Satellite Imagery and Dependency Profiles 96

Advanced Machine Learning-based Fifth-Generation Network Attack Detection System 98

LDRD OVERVIEW

INL's LDRD Portfolio

In 1991, DOE with the approval of the US Congress established the LDRD program as a means for the laboratories to devote a relatively 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. Truly, LDRD serves as a proving ground for advanced research and development concepts that may not have been otherwise pursued through direct programmatic funding sources.

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 four 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. Funds are available throughout the year to start these short-term, dollar-limited projects.
- 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 supports mid to late career researchers, providing opportunities to advance their scientific leadership while advancing INL's mission.

DOE LDRD OBJECTIVES

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

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

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

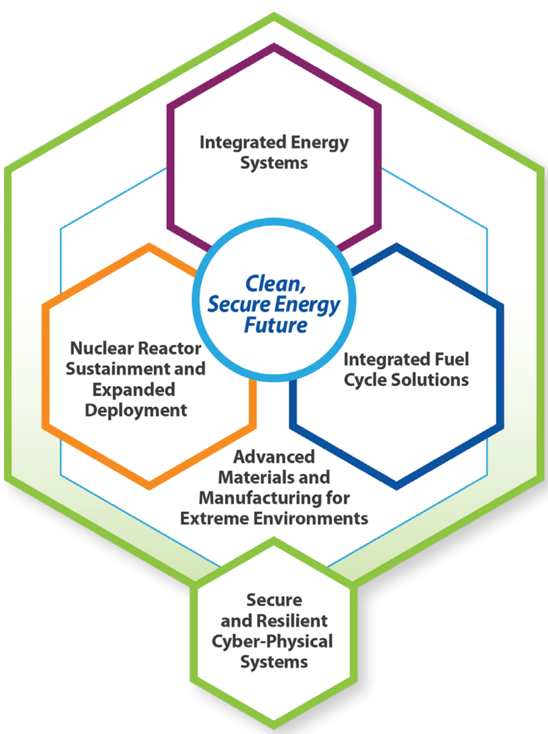
Core Capabilities

As of 2022, DOE recognized 24 core capabilities as foundational to its mission. Of those 24 capabilities, DOE acknowledges that INL has 13 core capabilities and 2 emerging capabilities. The acknowledgement of these capabilities highlights the exceptional breadth of INL's science and technology leadership that spans a continuum of basic and applied research and development.

INL HAS TWO EMERGING CAPABILITIES AND

13 CORE CAPABILITIES

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 two emerging core capabilities—Chemical and Molecular Science and Condensed Matter Physics and Materials Science—as strategic scientific initiatives. Of the 45 projects highlighted in this report, 23 demonstrated one or both emerging core capabilities.



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 requested on each proposal and project continuing to the next fiscal year prior to project funding.

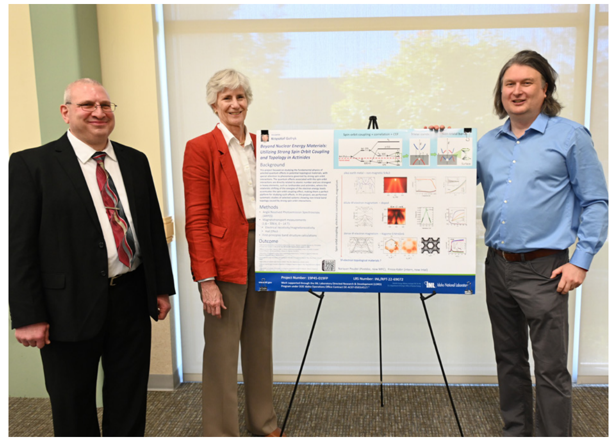
Showcasing Success

On September 19, 2022, INL hosted a poster session showcasing the LDRD projects ending in fiscal year 2022. This was the first in-person LDRD poster session since 2019. Attendees included INL researchers, industry and academic partners, external collaborators, and members of the public. Images of the posters are available on INL.gov.

Core capabilities

- Accelerator science and technology
- Advanced computer science, visualization, and data*
- Applied materials science and engineering*
- Applied mathematics
- Biological and bioprocess engineering*
- Biological systems science
- Chemical and molecular science**
- Chemical engineering*
- Climate change sciences and atmospheric science
- Computational science
- Condensed matter physics and materials science**
- Cyber and information sciences*
- Decision science and analysis*
- Earth systems science and engineering
- Environmental subsurface science*
- Large-scale user facilities /R&D facilities/advanced instrumentation*
- Mechanical design and engineering*
- Nuclear and radio chemistry*
- Nuclear engineering*
- Nuclear physics
- Particle Physics
- Plasma and future energy sciences
- Power systems and electrical engineering and integration*
- Systems engineering and integration*

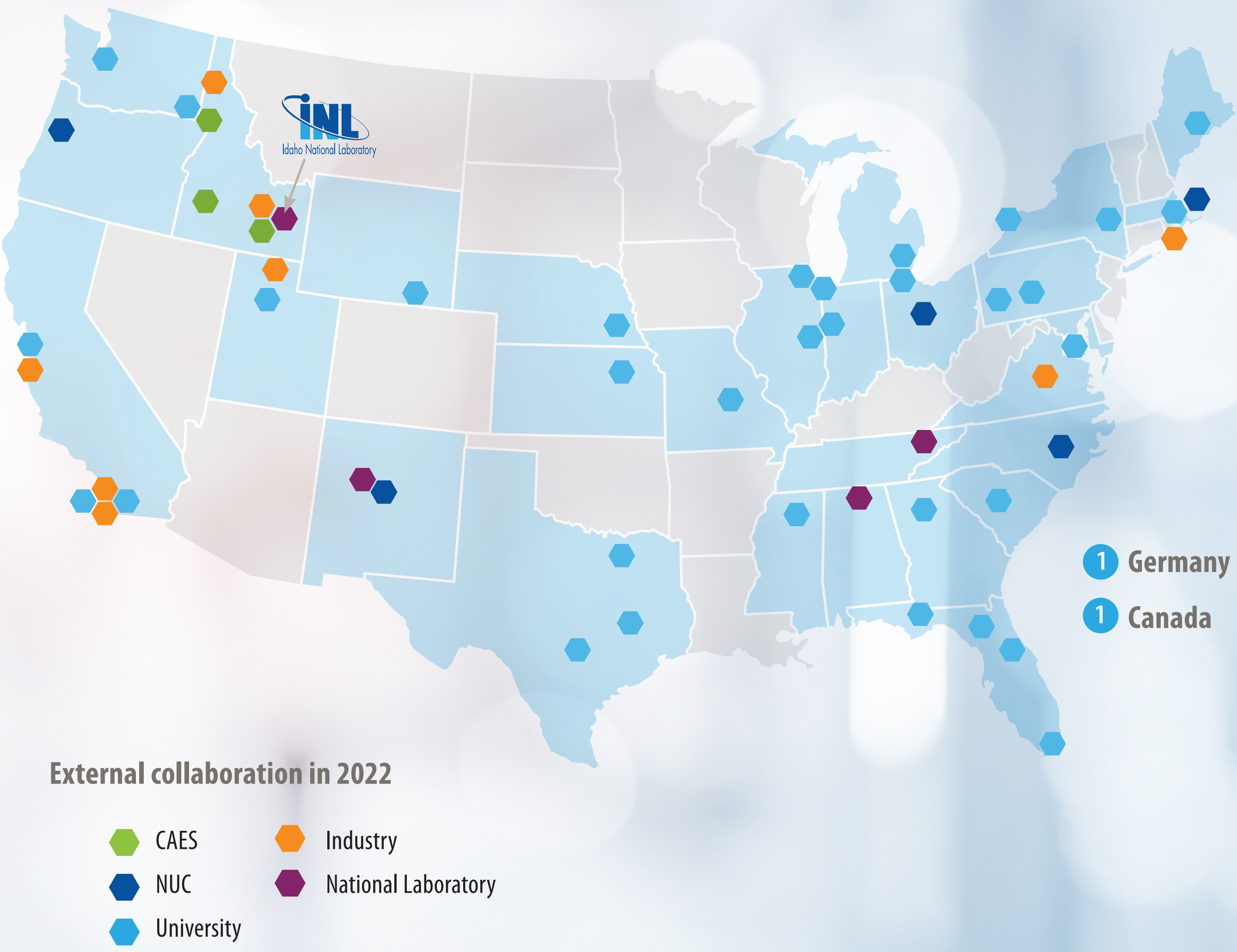
*Acknowledged ** Emerging



Dr. Krzysztof Gofryk won first place best poster at this year's annual poster session. Dr Gofryk's project, "Beyond Nuclear Energy Materials: Utilizing Strong Spin-orbit Coupling and Topology in Actinides," is an example of basic science advancing INL's mission.

Dr. Peter Zalupski won second place best poster at this year's annual poster session. Dr. Zalupski's Chemical and Molecular Science and Nuclear and Radiochemistry research advances cancer diagnosis and radiation treatment.

Dr. Geoffrey Beausoleil won third place best poster at this year's annual poster session. The team accomplished the first fabrication of actinide bearing multi-principal element alloys that could be used for high temperature reactor fuel.



111

RESEARCHERS OUTSIDE OF INL COLLABORATED ON LDRD PROJECTS

Collaboration

INL scientists and engineers collaborate with researchers around the world to advance the frontiers of science, technology, and engineering. In fiscal year 2022, 111 researchers outside INL collaborated on LDRD projects, including researchers from 28 states as well as Canada and Germany. Collaborative partnerships are primarily with universities, but also include industry and other national laboratories.

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

10 Graduate fellows from 9 institutions

43 Postdocs from 30 institutions

61 Interns from 34 institutions

97 University collaborators

14 Nonprovisional U.S. patent applications

19 Invention disclosure records

9 Patents granted

2 Copyright asserted

92 Publications

15 Software disclosure records

36 Licenses issued

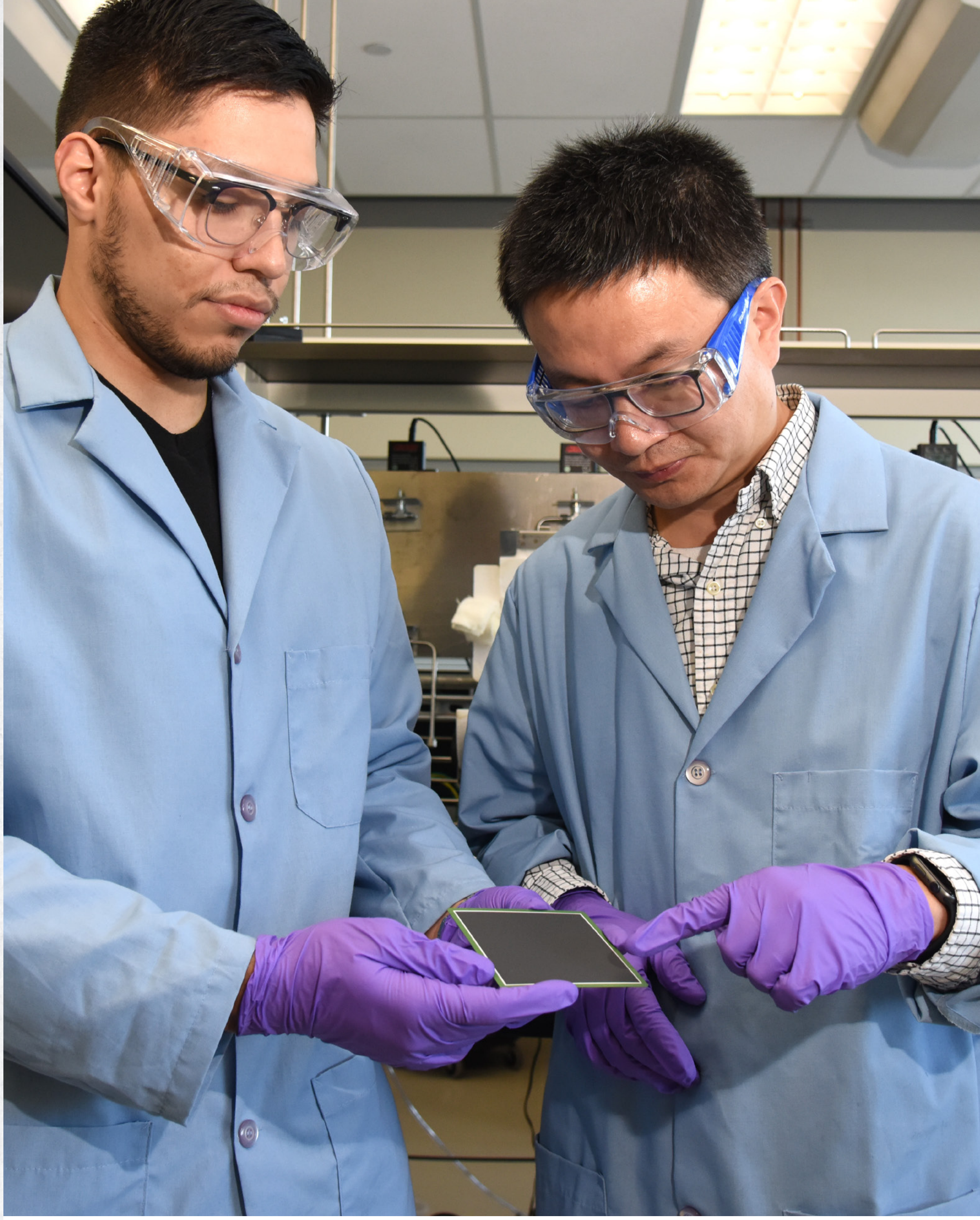
56 New projects

45 Projects ending

133 Active projects

8 Industry collaborators

6 DOE or other federal lab collaborators



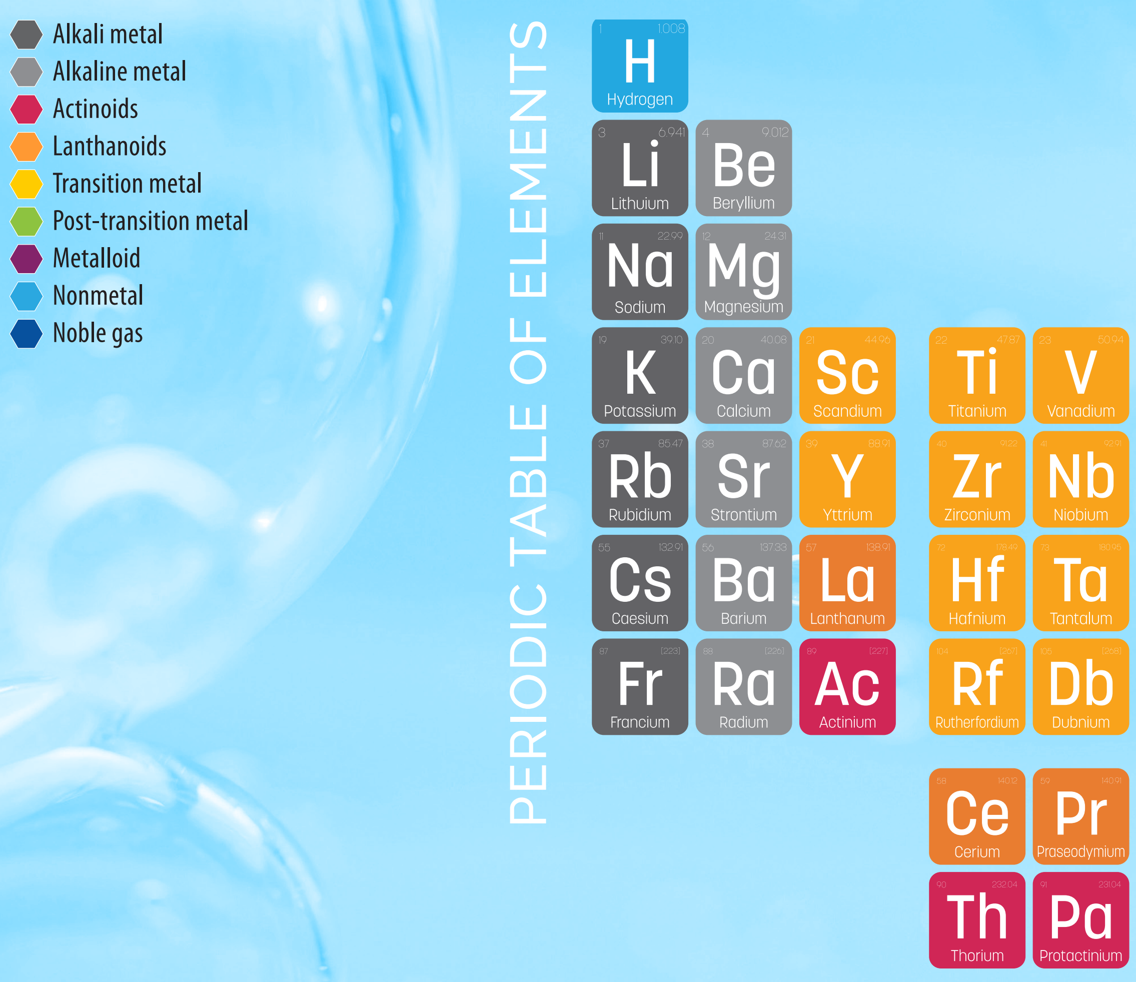
University, The Ohio State University, Oregon State University and University of New Mexico. Thirty-five researchers from these consortia were co-investigators on LDRD projects in fiscal year 2022, and many more students and postdocs contributed. Additionally, 62 researchers from other universities were co-investigators along with students and postdocs contributing.

$40

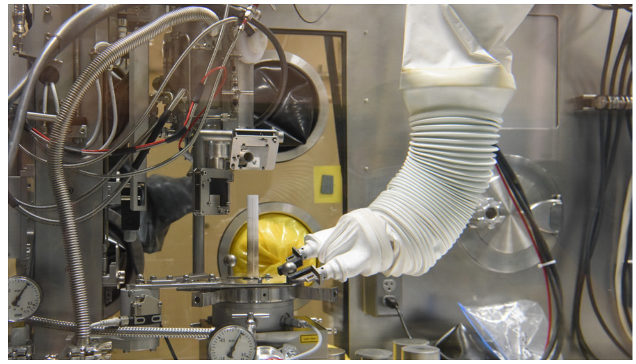
MILLION TOTAL PROJECT COST

ACRONYMS & ABBREVIATIONS

AM	additive manufactured
ARCTIC	Advanced fast Reactor Concept in Thermal-spectrum Irradiation Capability
DSC	differential scanning calorimetry
DOE	Department of Energy
FEM	finite element model
FIB	focused ion beam
IASCC	irradiation-assisted stress corrosion cracking
INL	Idaho National Laboratory
ISOPOD	inverse optimization and design
LDRD	Laboratory Directed Research and Development
LTE	Long Term Evolution
MOOSE	Multiphysics Object-Oriented Simulation Environment
MSR	molten salt reactor
NMC	nickel manganese cobalt
PCT	Patent Cooperation Treaty
RCIC	reactor core isolation cooling

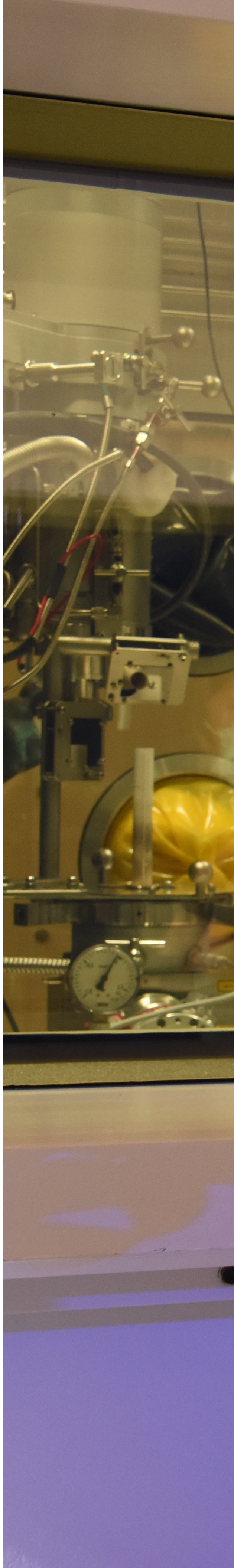


SCANN	smart contingency analysis neural network
SIMS	secondary ion mass spectroscopy
SPOCK	Spectral Observation Convolutional Neural Network
SPS	spark plasma sintering
TAP	temporal analysis of products
TGS	transient grating spectroscopy
TREAT	Transient Reactor Test facility
U.S.	United States
YAG	yttrium aluminum garnet



NUCLEAR REACTOR SUSTAINMENT AND EXPANDED DEPLOYMENT

As DOE's nuclear energy laboratory, INL is creating and defining the next phase of nuclear energy. INL advances global competitiveness by sustaining and extending the safe and efficient operation of existing reactors and by pioneering advanced reactor technologies for future deployment. INL focuses research, development, and demonstration of innovative technologies to improve the performance of existing and future nuclear energy systems. Under this initiative, INL uses its core capabilities to strengthen the domestic commercial nuclear energy enterprise, enable United States technological leadership in global nuclear energy markets, and expand and deploy national nuclear energy strategic infrastructures.




DANGER
HIGH RADIATION AREA
ACCESS CONTROLS REQUIRED
DOSE RATES > 1 R/HR @ 30CM
Dosimeter, Supplemental Dosimeter and RWP Required for Entry
Area located inside the sample well(s). Contact Rad Con prior to opening.

Beyond Nuclear Energy Materials: Utilizing Strong Spin-orbit Coupling and Topology in Actinides

PROJECT NUMBER:
19P45-019

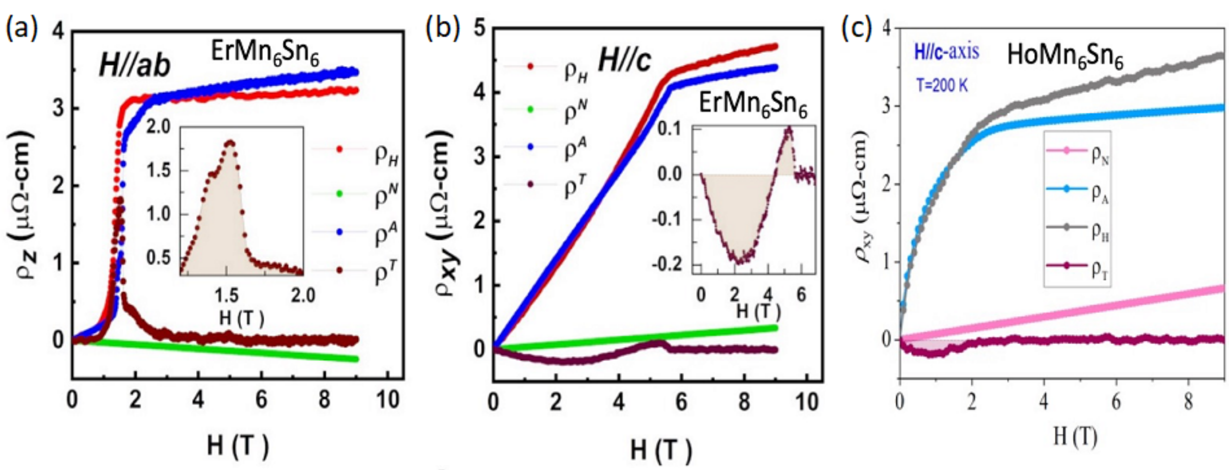
TOTAL APPROVED AMOUNT:
$525,500 over 3 years

PRINCIPAL INVESTIGATOR:
Krzysztof Gofryk

COLLABORATOR:
University of Central Florida

Detailed magnetotransport studies advance the fundamental understanding of spin-orbit interactions and topology in new topological insulators and semimetals.

This project focused on studying the fundamental physics of selected quantum effects in potential topological materials with special attention to phenomena governed by strong spin-orbit interactions. Topological phases, especially those governed by strong spin-orbit interactions, will be crucial for the realization of next generation quantum technologies. The quantum effects associated with the spin-orbit interactions are directly related to atomic number and are strongest in heavy elements, such as lanthanides and actinides, where the relativistic shifting of the energies of the electron energy levels accentuates the spin-orbit coupling effect, making them a perfect platform for studying such effects. In this project, selected systems showing non-trivial band topology caused by strong spin-orbit interactions were systematically studied. To better understand the interplay of many-body physics and other degrees of freedom, such as topology and electronic correlations, we performed detailed low temperature, magnetic, transport, thermodynamic, and spectroscopic studies of selected *s*, *p*, and *f*-electron materials. The project led to several high impact factor publications, and one of the papers was selected as the Top 100 Scientific Reports in Physics papers in 2020. Advancements in quantum materials research performed in this project supported recent lab initiatives toward condensed matter physics and INL's Center for Quantum Actinide Science and Technology.



(a-b) Anomalous and topological Hall effect of erbium-manganese-tin ($ErMn_6Sn_6$) at 200 K. Inset shows enlarged view of ρT vs. magnetic field (H). (c) Anomalous and topological Hall effect of holmium-manganese-tin ($HoMn_6Sn_6$) at 200 K.

TALENT PIPELINE:

- Firoza Kabir, student at the University of Central Florida
- Narayan Poudel, postdoc at the University of Houston, hired as staff at INL

PUBLICATIONS:

Mofazzel Hosen, M., G. Dhakal, B. Wang, N. Poudel, K. Dimitri, F. Kabir, C. Sims, S. Regmi, K. Gofryk, D. Kaczorowski, A. Bansil, and M. Neupane, "Experimental observation of drumhead surface states in $SrAs_3$," *Nature's Scientific Reports 10* (2020) 2776.

Mandujano, H. C., S. L. Gonzalez, N. Episcopo, U. Sitharaman, N. Poudel, K. Gofryk, Y. E. Garay, J. A. Lopez, Z. Qiang, S. Calder, H. S. Nair, "Absence of long-range magnetic order in lithium-containing honeycombs in the Li–Cr–Sb(Te)–O phases," *Journal of Physics: Condensed Matter* 33(29) (2021).

Kabir, F., Md. M. Hosen, X. Ding, C. Lane, G. Dhakal, Y. Liu, K. Dimitri, C. Sims, S. Regmi, A. P. Sakhya, L. Persaud, J. E. Beetar, Y. Liu, M. Chini, A. K. Pathak, J.-X. Zhu, K. Gofryk, M. Neupane, "Observation of topological surface state in gadolinium-doped Sb_2Te_3," *Frontiers in Quantum Materials (Quantum Materials)* 8 (2021) 706658.

Dhakal, G., F. C. Kabeer, A. K. Pathak, F. Kabir, N. Poudel, R. Filippone, J. Casey, A. P. Sakhya, S. Regmi, C. Sims, K. Dimitri, P. Manfrinetti, K. Gofryk, P. M. Oppeneer, M. Neupane, "Anisotropically large anomalous and topological Hall effect in a kagome magnet," *Physical Review B* 104 (2021) L161115.

S. Regmi, G. Dhakal, F. C. Kabeer, F. Kabir, C. Sims, Y. Liu, K. Dimitri, L. Persaud, N. Harrison, K. Gofryk, D. Kaczorowski, P. M. Oppeneer, M. Neupane, "Observation of multiple nodal lines in SmSbTe," *Physical Review Materials* 6 (2022) L031201.

Kabir, F., R. Filippone, G. Dhakal, Y. Lee, N. Poudel, J. Casey, A. P. Sakhya, S. Regmi, C. Sims, R. Smith, P. Manfrinetti, L. Ke, K. Gofryk, M. Neupane, A. K. Pathak, "Unusual magnetic and transport properties in $HoMn_6Sn_6$ kagome magnet," *Physical Review Materials* 6 (2022) 064404.

AWARD:

Hosen, M. M., G. Dhakal, B. Wang, N. Poudel, K. Dimitri, F. Kabir, C. Sims, S. Regmi, K. Gofryk, D. Kaczorowski, A. Bansil, M. Neupane, "Experimental observation of drumhead surface states in $SrAs_3$," Nature's Scientific Reports 10 (2020) 2776. The paper was selected as one of the Top 100 Scientific Reports physics papers in 2020.

Demonstrate Viability of Accelerated Fuel Qualification Approaches

PROJECT NUMBER:
20A44-036

TOTAL APPROVED AMOUNT:
$1,299,745 over 3 years

PRINCIPAL INVESTIGATOR:
Seongtae Kwon

CO-INVESTIGATORS:
Adrian Wagner, INL
Christopher Turner, INL
Geoffrey Beausoleil, INL

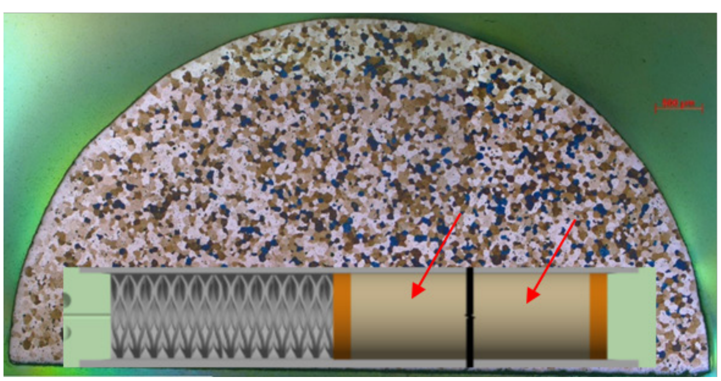
COLLABORATOR:
General Atomics

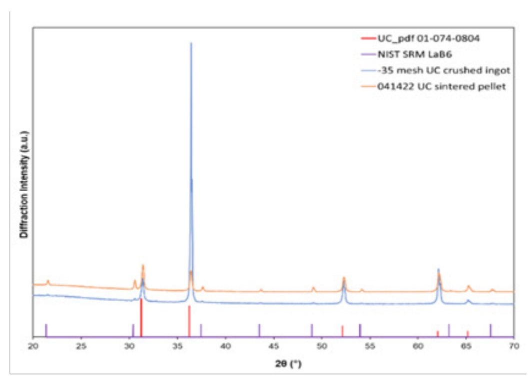
Expanding the Fission Accelerated Steady-state Testing approach to uranium carbide fuel qualification can drastically reduce the time and effort for nuclear fuel qualification.

The two leading contenders in accelerated fuel system qualification are INL's Fission Accelerated Steady-state Testing (FAST) and General Atomics' and Oak Ridge National Laboratory's Accelerated Fuel Qualification. Accelerated Fuel Qualification starts small, irradiating large numbers of spherical fuel particles under diverse conditions to develop inputs for mechanistic fuel behavior models. The FAST approach irradiates a sizable number of scaled down integral fuel pins to accelerate burn up by a factor of up to ten and is more suited for characterizing fuel performance at engineering-relevant length scales. These two approaches, while sufficiently differentiated to stand on their own, are highly synergistic. This presents an opportunity to demonstrate how the combined use of both approaches offers benefits that exceed either individually and how to use the best attributes of both concepts.

This research intended to test the hypothesis that fuel qualification is accelerated using the FAST approach. The approach is prototypic in some respects (e.g., peak temperature is maintained) but non-prototypic in others (e.g., temperature gradient is different). This project produced test articles for a ceramic fuel system that, based on analyses and specimen design conducted, provided data to demonstrate the viability of the approach for a range of fuel forms.

The fabrication process of uranium carbide fuel was developed and neutronic, thermal, and hydraulic analysis was conducted for FAST. The results provide the basis of expanding FAST as an efficient platform to broad nuclear fuels.





Fabricated uranium carbide pellets with 95% sintered density and schematic of test capsule assembly with fabricated fuel (left) and x-ray diffraction pattens of ingot and sintered pellets demonstrating phase pure uranium carbide phase.

Development of Novel Radiometal Chelators for Imaging and Therapeutic Nuclear Medicine

PROJECT NUMBER:
20A44-089

TOTAL APPROVED AMOUNT:
$1,100,000 over 3 years

PRINCIPAL INVESTIGATOR:
Peter Zalupski

CO-INVESTIGATORS:
John Klaehn, INL
Aidan Bender, University of Utah
Tara Mastren, University of Utah
Santa Jansone-Popova, Oak Ridge National Laboratory

Linking a multi-functional molecular construct to a tumor-seeking antibody enables cancer diagnosis and radiation treatment.

Energy of radioactive decay may be deposited to the malignant cell if an unstable atom is delivered near a tumor. This well-known radiotherapeutic concept was a backbone of this project where a multi-modal radiometal carrier was built for numerous biomedical applications. The molecular unit uses a functionalized phosphazene platform, which is decorated with structures designed to bind radiometals and attach to cancer-seeking antibodies. This approach adds versatility to diagnostics and treatment of cancer with radiation. Different radiometals can be delivered near tumor cells to offer concerted options for positron emission tomography, Cherenkov-sensitized fluorescence, magnetic resonance imaging, and x-ray induced Auger electron radiosensitization.

Novel molecules (chelates) were synthesized to build strong coordination environments and withstand metal binding pressures of complex in vivo conditions. Chelates coordinate a variety of medical isotopes such as radiostrontium and radiolanthanides, serving as "armored delivery vehicles" in the human body. In this work, chelates were adapted to offer compatibility with "click" chemistry, which is a convenient bioconjugation tool for linking molecular units. This synthetic strategy connected multiple chelates to a phosphazene platform. Bioconjugation, i.e., attachment of chelate to antibody, was also enabled via "click" chemistry after antibody surface functionalization.

TALENT PIPELINE:
Aidan Bender, student at University of Utah

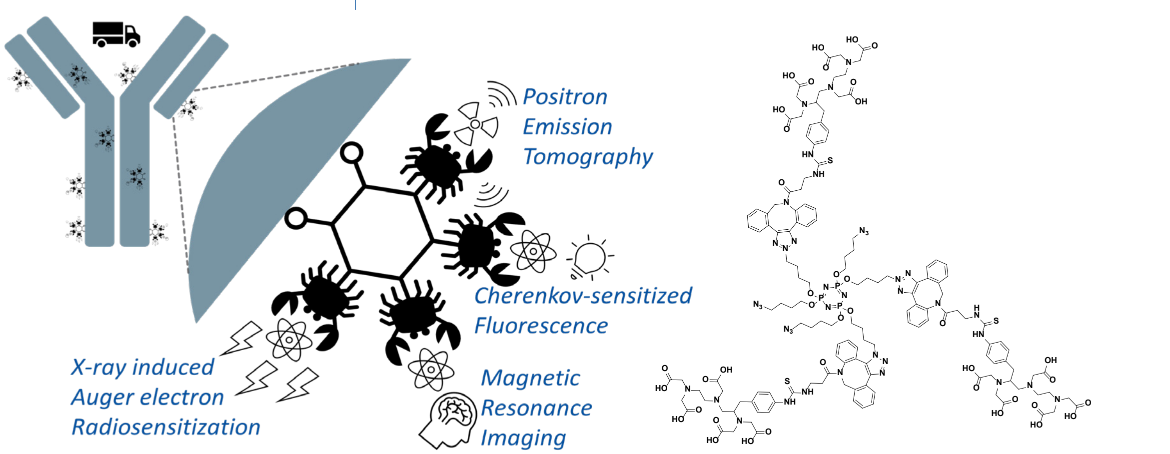


Illustration of multi-modal radiometal carrier concept developed for simultaneous tumor imaging and therapy options. Structure shows a synthesized molecular construct where three chelate units are available for radiometal binding and three vectors are available for attachment to monoclonal antibody.

Combinatorial and High Throughput Materials Synthesis to Advance Nuclear Materials Discovery

PROJECT NUMBER:
20A44-046

TOTAL APPROVED AMOUNT:
$1,486,000 over 3 years

PRINCIPAL INVESTIGATOR:
Jason Schulthess

CO-INVESTIGATORS:
Evander Evans Chambers, INL
Thomas Maddock, INL
Frank Liou, Missouri University of Science and Technology
Joseph Newkirk, Missouri University of Science and Technology

COLLABORATORS:
CalNano, Inc.
Central Valley Machine

Demonstrated proof-of-principle blazes trail for computational materials engineering tools to help nuclear structural materials discovery and to reduce time and cost for irradiations in the Advanced Test Reactor.

Researchers developed and demonstrated combinatorial workflows for structural materials discovery and neutron irradiation testing of structural materials. Using modern computational materials engineering tools to explore the complex composition space provided by the class of alloys known as multi-principal element alloys. Multiple compositions were selected for rapid fabrication in a fast-to-fail approach. This approach avoided the historical and time intensive iterative approach and allowed for rapid down selection of candidate materials. Samples were also fabricated by both spark plasma sintering and arc melting to provide further comparison of rapid fabrication techniques. Baseline characterization of the candidate materials selected and fabricated confirmed modeling predictions, thus validating the use of the modeling tools for alloy design and initial selection. The presence of deleterious brittle phases, extremely high hardness, and poor ductility were found in some of the initially selected alloys. The modeling tools were then used to modify the compositions to obtain a softer major phase with a dispersed hard phase to enhance mechanical properties while maintaining the potential for high temperature creep resistance.

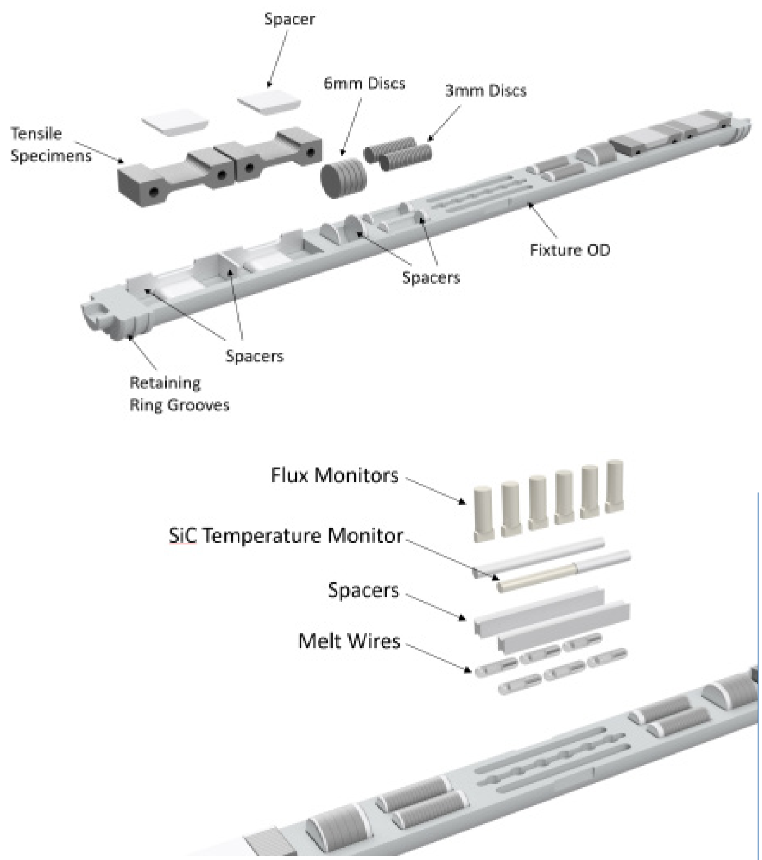
A standard capsule for structural materials irradiation in the Advanced Test Reactor (ATR) was developed and demonstrated by irradiating 240 specimens from 16 material compositions selected during the initial material discovery phase. While structural materials testing has previously occurred in ATR, the capsule design and fabrication process usually takes years to complete. To decrease the cost and time associated with future materials testing in ATR, the standard capsule was designed to hold multiple specimen geometries and allow use in virtually any ATR irradiation position. The standard capsule features locking endcaps that connect and lock together to form the capsule stack, eliminating the need for a basket and maximizing the quantity of specimens that are contained in the capsule. A thermal model was developed that requires only minimal changes to update the material properties and heating rates for new experiments. A key outcome is two additional irradiation experiments that are using the standard capsule design.

TALENT PIPELINE:

- Anilas Karimpilakkal, student at Missouri University of Science and Technology
- Cesar Ortiz Rios, student at Missouri University of Science and Technology
- Marcus Parry, student at University of Utah
- Sriram Praneeth Isanaka, student at Missouri University of Science and Technology

PUBLICATIONS AND PRESENTATIONS:

Karimpilakkal, A., J. Newkirk, F. Liou, J. S. S. P. Isanaka, J. Medvedeva, "Modelling of equiatomic Mo-Nb-Ti-Zr and Mo-Nb-Ti systems for use in radiation environments," *High Entropy Alloys 2021 Conference*, Charlotte, NC, USA, Dec. 5–8, 2021.



Schematic of standard capsule showing specimens and in situ instrumentation. (SiC – silicon carbide)

Real-time Axial Neutron Flux Profile Measurement at the Advanced Test Reactor Critical Facility

PROJECT NUMBER:
20A44-112

TOTAL APPROVED AMOUNT:
$824,000 over 3 years

PRINCIPAL INVESTIGATOR:
Michael Reichenberger

CO-INVESTIGATOR:
Mary Rose Holtz, INL

New sensor testing capabilities enable real-time neutron flux monitoring at the Advanced Test Reactor Critical facility.

Real-time characterization of irradiation facilities improves the utilization of the core capabilities of test nuclear reactors. The ability to observe how the local neutron flux (level and spectrum) changes as control elements and experiments change will fundamentally transform our understanding of the underlying physical phenomena that govern the operation of present and advanced nuclear reactors, ultimately providing valuable information for the nuclear energy industry. The objective of this research was to demonstrate how advanced sensors could be used to significantly reduce the time and cost of experiments, improve our understanding of experimental environments, and enable verification and validation of simulation and modeling methods. This was accomplished by designing and fabricating a dedicated real-time instrument test train for the ATR Critical facility.

The first year of this project focused on the design and modeling of real-time axial neutron flux monitors, leveraging proven technologies pioneered at INL, to characterize the transient that occurs in the small 'B' positions at ATR and the ATR Critical facility. We found that the flux amplitude in those positions can fluctuate as much as 380% depending on the outer shim control cylinder position. These fluctuations create large uncertainties for experiment designs that lead to substantial deviations between simulation results and as-run measurements.

The engineering design of the test fixture and flux monitor instrumentation was the objective of the second project year. Most experiments at ATR and ATR Critical facility require unique test fixture fabrication for the irradiation position within the reactor. This project was no different; however, a keen focus was given to the flexibility of this test fixture to facilitate multiple real-time instruments both present and future. New capabilities were established to electrodeposit enriched uranium for fission chamber development at INL, and trials were begun to characterize the process. Substantial work was also necessary to formalize the instrument design process as well. The test fixture and instrument designs were finalized, reviewed, and released so that the fabrication of the Quality Level 3 components could be initiated.

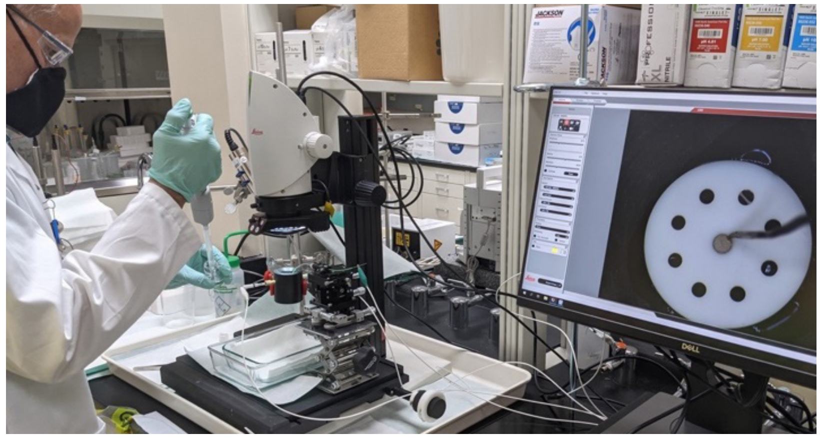
The final year included the fabrication of the test fixture and instruments for ATR Critical facility. This unique capability that we delivered to ATR Critical facility will facilitate future instrument testing and scientific experiments.

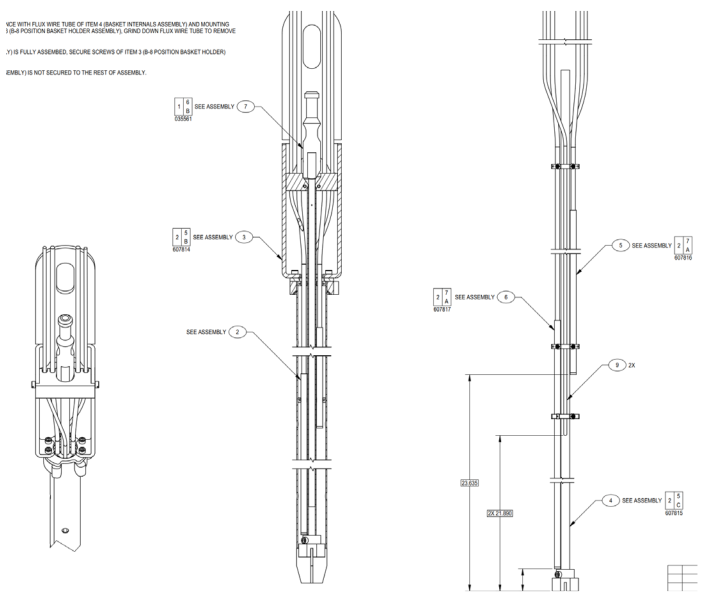
TALENT PIPELINE:

- Mary Rose Holtz, student at University of Idaho
- Dan Nichols, student at Kansas State University

PUBLICATION:

Nichols, D. M., M. A. Reichenberger, A. D. Mailie, M. R. Holtz, D. S. McGregor, "Simulated Performance of the Micro-Pocket Fission Detector in the Advanced Test Reactor Critical Facility," *Nuclear Science and Engineering* 195(10) (2021) 1098–1106.





(top) New electrodeposition techniques were implemented at the ATR Radioanalytical Chemistry Laboratory to enhance fission chamber fabrication capabilities at INL. (bottom) The assembled test fixture was completed at the ATR Test Train Assembly Facility.

Modeling and Characterization of Alpha-uranium to Accelerate Metallic Fuels Development

PROJECT NUMBER:
20A44-121

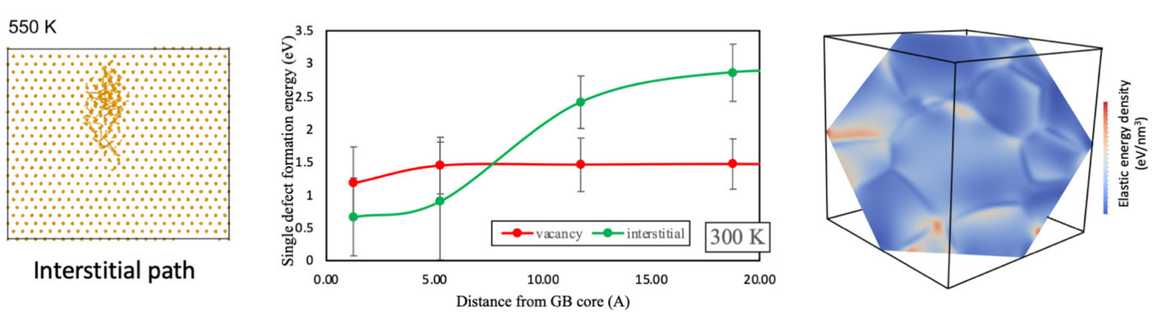
TOTAL APPROVED AMOUNT:
$1,755,109 over 3 years

PRINCIPAL INVESTIGATOR:
Andrea Jokisaari

CO-INVESTIGATORS:
Cheng Sun, INL
Michael Benson, INL
Xiang Liu, INL
Benjamin Beeler, North Carolina State University

Understanding the fundamental irradiation-induced microstructure and property changes of alpha-uranium enables metallic fuel research and development and basic energy science.

This project focused on the investigation of irradiation-induced microstructure and property changes in alpha-uranium via a combination of modeling and experimental techniques. Alpha-uranium is the low temperature phase of uranium and is present in certain metallic fuel alloys. The heyday of alpha-uranium research was in the 1960s, prior to the invention and widespread availability of many modern materials science techniques. As a result, a fundamental understanding of the physics governing the irradiation response of alpha-uranium is lacking. Due to the strongly anisotropic nature of alpha-uranium, which is driven by its orthorhombic crystal structure, an understanding of the behavior cannot simply be projected from other metals with similar physical properties. The project elucidated basic physical properties to understand fundamental aspects of irradiation behavior of alpha-uranium. The focus was primarily on the behavior of point defects, their transformation to extended defects, and the effect of interfaces on irradiation tolerance and other mechanical behaviors and properties. This helped build basic science capabilities for irradiation research and aid physics-based fuel performance models development. A combination of experiment and atomistic and mesoscale modeling were used in this work. This research greatly expanded the knowledge of the fundamental properties of alpha-uranium, shed light on factors controlling the complex irradiation response of the material, provided information useful to thermomechanical processing of the material, and developed a new in situ furnace for the Neutron Radiography Reactor at INL. These results add to the basic science capability of INL and improve the ability to fabricate and model certain metallic fuels. This project also improved integration between modeling, experimental, and engineering capabilities at INL and strengthened collaboration between INL and North Carolina State University.



Conceptual illustration of the multi-scale mechanistic approach to studying the behavior of alpha-uranium. Left) The path of a diffusing interstitial at 550 K simulated with molecular dynamics. Middle) A measure of the sink strength of a grain boundary for vacancies and interstitials simulated with molecular dynamics. Right) The spatially-dependent elastic energy density in a polycrystalline alpha-uranium sample.

TALENT PIPELINE:

- Arunkumar Seshadri, postdoc at INL
- Khadija Mahbuba, student at North Carolina State University
- Yuhao Wang, student at University of Michigan

PUBLICATIONS AND PRESENTATIONS:

A. Seshadri, A. M. Jokisaari, C. Sun, "A review of irradiation damage and effects in a-uranium," *Materials* 15 (2022) 4106.

Jokisaari, A. M., K. Mahbuba, Y. Wang, B. Beeler, "The impact of anisotropic thermal expansion on the isothermal annealing of polycrystalline a-uranium," *Computational Materials Science* 205 (2022) 111217.

Mahbuba, K., B. Beeler, A. Jokisaari, "Evaluation of the anisotropic grain boundaries and surfaces of a-U via molecular dynamics," *Journal of Nuclear Materials* 544 (2021) 153072.

Beeler, B., K. Mahbuba, Y. Wang, A. Jokisaari, "Determination of thermal expansion, defect formation energy, and defect-induced strain of a-U via ab initio molecular dynamics," *Frontiers in Materials* 8 (2021) 188.

Mahbuba, K., "Grain boundary energy and surface energy of alpha-uranium," *American Nuclear Society Student Conference* (2021).

Jokisaari, A., "The challenges of alpha-uranium: Fundamental Understanding of a Past and Future Nuclear Fuel Material," *Materials in Nuclear Energy Systems* (2021) (Invited talk).

Wang, Y., B. Beeler, A. Jokisaari, "An atomistic study of the anisotropic elastic response of defects in alpha uranium," *The Minerals, Metals and Materials Society* (2022).

Mahbuba, K., B. Beeler, A. Jokisaari, "Atomistic modeling of transport properties and interaction with point defects of α-U tilt grain boundaries," *The Minerals, Metals and Materials Society* (2022).

Development of a Streamlined Approach for Burnup and Microstructural Analysis of Nuclear Materials from Nanoscale to Mesoscale

PROJECT NUMBER:
20A44-153

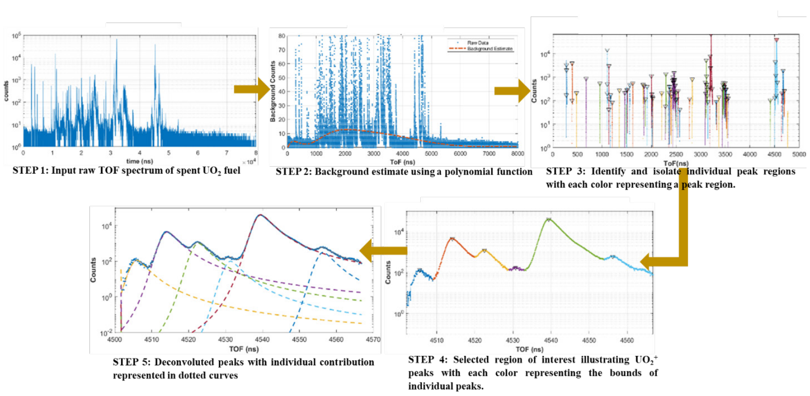
TOTAL APPROVED AMOUNT:
$1,000,000 over 3 years

PRINCIPAL INVESTIGATOR:
Mukesh Bachhav

CO-INVESTIGATORS:
Brandon Miller, INL
Daniel Wachs, INL
Dennis Keiser, INL
Jian Gan, INL
Joey Charboneau, INL
Joshua Kane, INL
Laura Sudderth, INL
Lingfeng He, INL
Matthew Jones, INL
Nick Erfurth, INL

State-of-the-art nuclear material profiling capabilities from nanoscale to mesoscale enable fundamental research of nuclear material behavior.

Pre- and post-irradiation analysis of nuclear materials can be an expensive and time-consuming endeavor due to the unavoidable processes necessary to gather accurate and precise results. An analysis can take anywhere from a few days to multiple weeks depending on the number of samples, activity of the sample, complexity of the fuel type, and measurement requests. Conventional methods involve surface-based measurements, such as scanning electron microscopy, and destructive analytical techniques, such as inductively coupled plasma-mass spectrometry. While these methods have their inherent advantages, they also suffer from the inability to rapidly acquire local isotopic information about the fuel sample. This effectively limits the information that can be derived about material performance. These factors prompt the need for an updated approach that improves upon the pitfalls of traditional methods while making notable advances in the analysis of nuclear materials. By merging the unique capabilities of atom probe tomography with a femtosecond laser ablation (LA)-laser induced breakdown spectroscopy (LIBS)-time-of-flight mass spectrometry (TOFMS), combined as LA-LIBS-TOFMS, this research increased sample throughput and provided a new method to quantitatively evaluate material performance based on three-dimensional isotopic distributions in a specimen. Atom probe tomography offers the unrivaled capability to quantitatively analyze for isotopic composition on a three-dimensional atomic scale. However, finding the most significant location to analyze can be a burdensome process. LA-LIBS-TOFMS offers many advantages that can complement atom probe tomography. Through rapid elemental and isotopic mapping of the samples surface at micron resolution, LA-LIBS-TOFMS improves upon customary scanning electron microscopy analysis used to identify locations for focused ion beam sampling.
To enhance the complementary capabilities of these techniques, we validated a quantitative protocol for LA-LIBS-TOFMS that can be correlated to atom probe tomography data. Our research improved shortcomings of each method through a joint approach, leaning on the concomitant nature of the techniques.



Workflow developed for atom probe tomography mass spectrum using MATLAB® toolbox for quantifying burnup from spent fuel.

TALENT PIPELINE:

- Anshul Kamboj, student at University of Michigan
- Olivia Licata, student at University at Buffalo
- Megan Burrill, student at Illinois Institute of Technology
- Lashavio Little, student at Prairie View A&M

PUBLICATIONS AND PRESENTATIONS:

Bachhav, M., B. Miller, J. Gan, D. Keiser, A. Leenaers, S. V. Berghe, M. K. Meyer, "Microstructural Changes and Chemical Analysis of Fission Products in Irradiated Uranium-7 wt.% Molybdenum Metallic Fuel Using Atom Probe Tomography," *Applied Sciences* 11(15) (2021) 6905.

Bachhav, M., J. Gan, D. Keiser, J. Giglio, D. Jädernäs, A. Leenaers, S. V. Berghe, "A novel approach to determine the local burnup in irradiated fuels using Atom Probe Tomography (APT)," *Journal of Nuclear Materials* 528 (2020) 151853.

Bachhav, M., J. Kane, F. Teng, F. Cappia, L. He, "Isotopic Analysis of Irradiated Ceramic Fuel for Burnup and Microchemical Assessment Using Atom Probe Tomography," *Microscopy and Microanalysis* 27(S1) (2021) 416–417.

Bachhav, M., L. He, J. Kane, X. Liu, J. Gan, F. Vurpillot, "Atom Probe Tomography for Burnup and Fission Product Analysis for Nuclear Fuels," *Microscopy and Microanalysis* 26(S2) (2020) 3086–3088 (Invited talk).

Yu, Z., M. Bachhav, F. Teng, L. He, A. Couet, "Nanoscale redistribution of alloying elements in high-burnup AXIOM-2 (X2®) and their effects on in-reactor corrosion," *Corrosion Science* 190 (2021) 109652.

Ditter, A. S., D. E. Smiles, D. Lussier, A. B. Altman, M. Bachhav, L. He, M. W. Mara, C. Degueldre, S. G. Minasian, D. K. Shuh, "Chemical and elemental mapping of spent nuclear fuel sections by soft x-ray spectromicroscopy," *Journal of Synchrotron Radiation* 29(1) (2021).

Bachhav, M., "Chemical and Microstructural Analysis of Nuclear Fuels at Nano-Length Scale Using Atom Probe Tomography," *AVS Conference*, (2021) (Invited talk).

Bachhav, M., "Elucidation Nuclear Materials Using Atom Probe Tomography," *Advanced Microscopy for Nuclear fuels, and Materials*, 2021 (Invited talk).

Mukesh Bachhav, Lingfeng He, Brandon Miller, Xiang Liu, Fabiola Cappia, Jian Gan, "Microstructural and Fission Products Analysis from Irradiated UO2 Fuel Using Atom Probe Tomography," *The Minerals, Metals and Materials Society conference*, Mar. 15–18, 2021.

Jones, M., "Quantitative Evaluation of U-Zr Alloy Fuels Utilizing Femtosecond LIBS," To be presented at *SciX 2022* (Invited talk).

Assessment of Irradiation Creep Using Accelerator Technology to Down-select Materials for In-pile Testing

PROJECT NUMBER:
20A44-155

TOTAL APPROVED AMOUNT:
$950,000 over 3 years

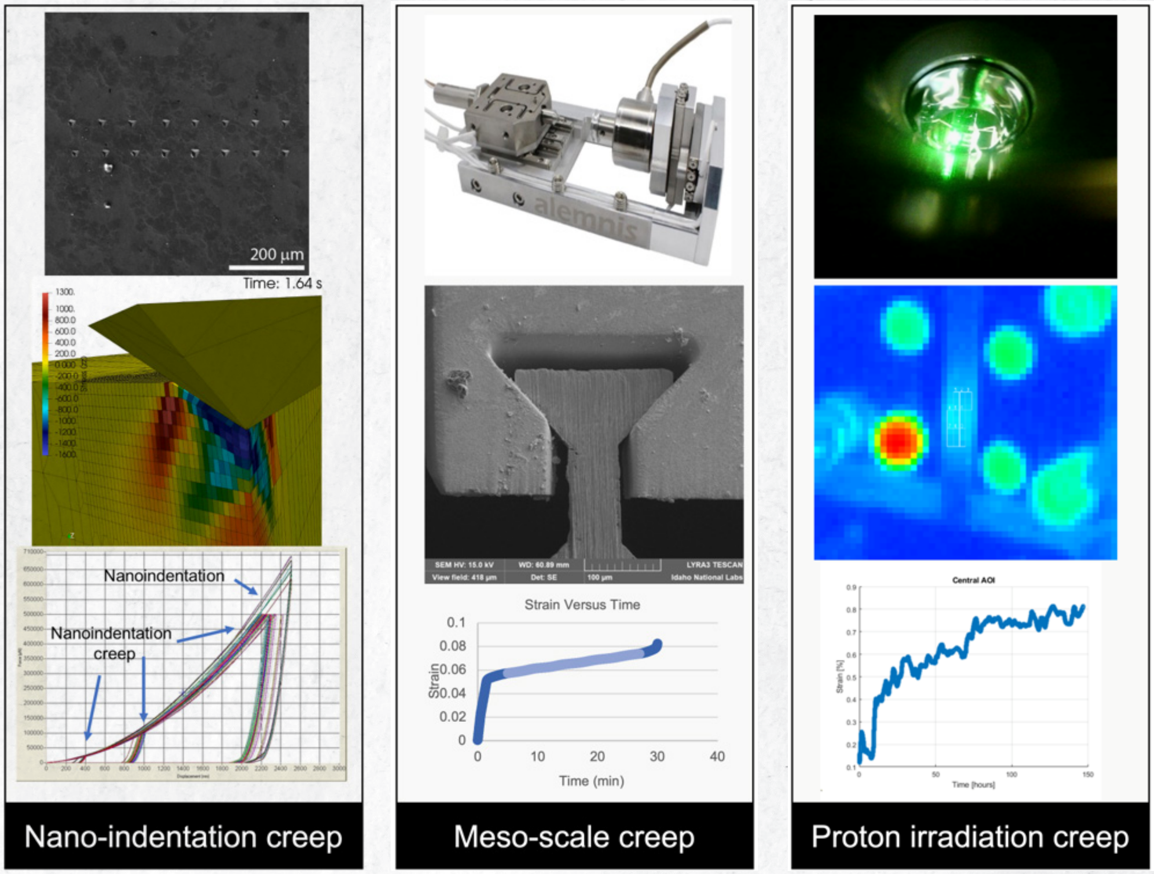
PRINCIPAL INVESTIGATOR:
Wen Jiang

CO-INVESTIGATORS:
Boopathy Kombaiah, INL
Cheng Sun, INL
Colin Judge, INL

COLLABORATOR:
University of Michigan

Small-scale thermal and irradiation creep measurements accelerate advanced materials development.

Advanced materials with enhanced resistance to irradiation at high temperatures benefit the life-extension and long-term operation of the current light water reactor fleet and the development of advanced nuclear reactor concepts. To qualify a material for reactor operation, many mechanical properties must be assessed, including thermal and irradiation creep. Creep is a time-dependent deformation process for a metal under the influence of stress, temperature, and irradiation. It is important to assess all aspects of the creep properties of new materials for reactor applications: thermal creep of as-manufactured materials, in-reactor/irradiation creep, and thermal creep of irradiated materials. In this project, accelerated creep measurement techniques were developed to assist in rapid qualification of new reactor materials. Those techniques include nanoindentation creep testing, mesoscale thermal creep testing and small-scale proton irradiation creep testing. In the nanoindentation testing, the creep of oxide-dispersion-strengthened stainless steel was performed and characterized. The nanoindentation modeling with Multiphysics Object-Oriented Simulation Environment (MOOSE) was used to extract the creep properties from the nanoindentation creep data. The novel mesoscale creep experiments on zirconium-niobium specimens with sizes of tens of micrometers were performed to extract bulk creep properties. Their measured rate of deformation shows good agreement to the bulk creep testing data. The in situ irradiation creep of 304 stainless steel was demonstrated using proton irradiation facilities at Michigan Ion Beam Laboratory. The irradiation temperature, strain, load, and proton beam current were recorded to obtain creep properties. This project paved a way to obtain creep properties of materials using only a small volume of specimen, facilitating efficient use of precious neutron-irradiated materials. This will significantly shorten the nuclear fuel and material development cycle and expedite the deployment of advanced materials for in-core applications.



Three accelerated small-scale creep measurement techniques: nanoindentation creep, mesoscale thermal creep, and combined thermal and proton irradiation creep.

TALENT PIPELINE:

- Marcus Parry, graduate fellow at INL
- Arunkumar Seshadri, postdoc at INL
- Mackenzie Warwick, student at University of Michigan
- Darren Parkison, student at University of California, Berkeley

PUBLICATIONS AND PRESENTATIONS:

Parry, M., "Ion and proton irradiations: In-situ experiments – microstructure, creep, and corrosion," Accelerated Irradiations for Reactor Structural Materials – *Virtual Meeting*, Idaho National Laboratory, Sep. 14–18, 2020.

Parry, M., C. Judge, C. Sun, W. Jiang, B. Kombaiah, G. Was, J. Aguiar, T. Sparks, "Accelerated study of thermal and irradiation creep in Fe-based multi-principal element alloys," *The Minerals, Metals and Materials Society 2021 Virtual Meeting*, Mar. 15–18, 2021.

Parry, M., B. Kombaiah, C. Judge, C. Sun, W. Jiang, O. Toader, G. Was, and T. Sparks, "Accelerated study of proton irradiation creep and hardening in Fe-Cr-Ni-based multi-principal element alloys," *2nd World Congress on High Entropy Alloys*, Charlotte, NC, USA, Dec. 4–8, 2021.

Develop High Fidelity Computation Models to Calculate the Effective Material Properties of Porous Cells

PROJECT NUMBER:
20A44-177

TOTAL APPROVED AMOUNT:
$1,448,000 over 3 years

PRINCIPAL INVESTIGATOR:
Mohammad Abdo

CO-INVESTIGATORS:
Boopathy Kombaiah, INL
Isabella Von Rooyen, INL
Yu-Lin Shin, University of New Mexico

COLLABORATORS:
Center of Advanced Energy Studies
The Ohio State University

New machine learning models can predict the effective thermal conductivity of closed-cell porous media, crack-containing microstructures, and additively manufactured materials.

The team developed machine learning metamodels that can predict the effective thermal conductivity of closed-cell porous media and crack-containing microstructures, such as tristructural isotropic compacts, and additively manufactured materials. This was accomplished by exploring the design space and perturbing all related features to identify the effect of pore size, crack size, orientation, geometric configuration, as well as tristructural isotropic particle distribution on effective thermal conductivity.

To achieve this goal, physics-based modeling through finite element analysis was performed, followed by a systematic machine learning pipeline covering data sampling, preprocessing, cleaning, sensitivity studies, surrogate modeling, then model validation. The model evolved from encompassing a single cell with a single spherical pore in a two-dimensional setup to multiple pores organized in several configurations to elliptical pores, to facilitate the control of the aspect ratios, and hence model cracks as well. Next, tristructural isotropic particles were introduced. Then the pores and tristructural isotropic particles were randomized following distributions fitted from the x-ray computed tomography scans of the available samples.

Several metamodels were constructed including linear regressors, support vector regressors, random forests, polynomial regressors, and feed-forward neural networks. These metamodels were validated and showed a coefficient of determination score that exceeded 0.95, reflecting the goodness of fit. However, these models still require the user to perform some data preprocessing to compute features of the geometric distribution. Hence, to render this more efficient, an ensemble model was built to concatenate the numeric data reflecting local conductivities, porosity, and particle packing fractions with a convolutional neural network to capture the geometric distribution, shapes, and orientations of pores and tristructural isotropic particles. This final model was in turn validated.

Finally, the model was converted to a three-dimensional finite element model. At each evolution, metamodels were built to assess the accuracy and compare to the literature if available. The research resulted in two journal articles for the sensitivity studies, one for the experimental analysis, and one for the machine learning work. The general finding was that the influential attributes were the local conductivities of the composite material and tristructural isotropic particles, the pore and crack densities, and the particle packing factor. Minor effects were observed and attributed to orientations and configurations. Insensitivity to orientation suggested the isotropic nature of the heat flux traveling through each medium.

TALENT PIPELINE:

- Luis Nuñez III, student at University of Idaho
- Calvin Downey, student at University of Idaho
- Kevin Irick, student at University of New Mexico
- Mahyar Pourghasemi, student at University of New Mexico
- Siavash Nikravash, postdoc at University of New Mexico

PUBLICATIONS AND PRESENTATIONS:

Shen, Y.-L., M. G. Abdo, I. J. V. Rooyen, "Numerical Study of Effective Thermal Conductivity for Periodic Closed-Cell Porous Media," Transport in Porous Media (2022).

Shen, Y.-L., M. G. Abdo, I. J. V. Rooyen, "Thermal Conductivity of Crack-Containing Media: A Numerical Study," *Journal of Composite Materials* 56 (2022) 2495-2508.

Kane, J. J., D. W. Marshall, N. L. Cordes, W. C. Chuirazzi, B. Kombaiah, I. v. Rooyen, J. D. Stempien, "3D analysis of TRISO fuel compacts via x-ray computed tomography," *Journal of Nuclear Materials* 565 (2022) 153745.

Mondal, K., L. Nuñez III, C. M. Downey, I. J. v. Rooyen, "Thermal barrier coatings overview: Design, manufacturing, and applications in high-temperature industries," *Industrial & Engineering Chemistry Research* 60(17) (2021) 6061–6077.

Shen, Y.-L., M. Abdo, B. Pham, I. v. Rooyen, "Effects of Void Configuration on the Overall Thermal and Mechanical Behavior of Porous Materials: A Numerical Modeling Approach," *The Minerals, Metals and Materials Society* 2021 Virtual, Mar. 15–18, 2021.

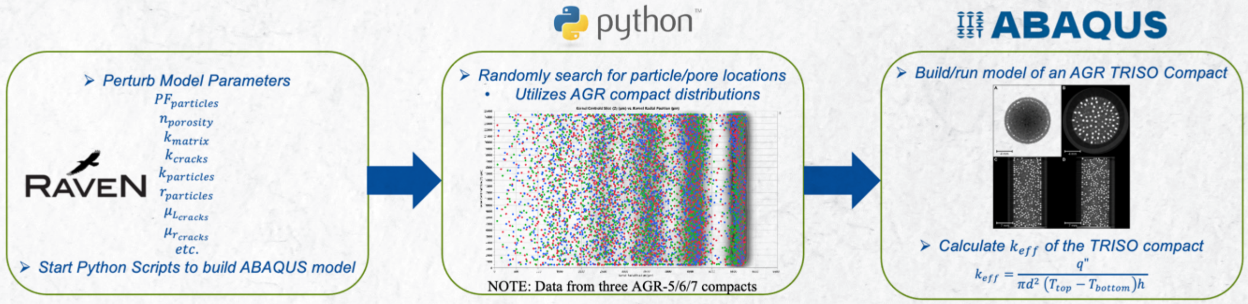
Abdo, M. G., Y.-L. Shen, I. J. v. Rooyen, "A Metamodel for Predicting Effective Thermal Conductivity of Porous Materials," *The Minerals, Metals and Materials Society 2021 Virtual*, Mar. 15–18, 2021.

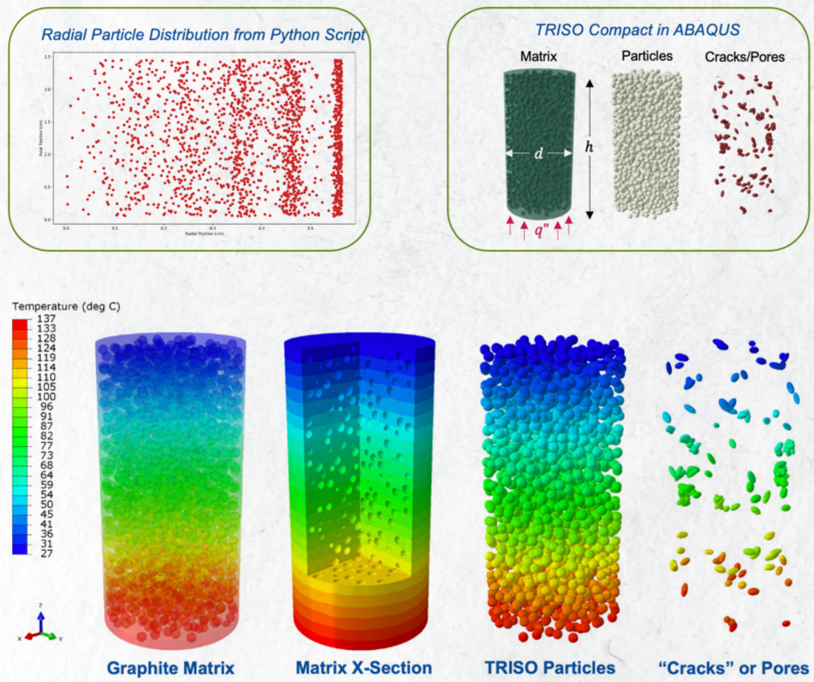
S. C., I. J. v. Rooyen, "Improved Techniques for Determining Local Thermal Transport in Composite Nuclear Fuels," *The Minerals, Metals and Materials Society 2021 Virtual*, Mar. 15–18, 2021.

Nuñez, L., I. v. Rooyen, "Experimental Fabrication of Porous Additive Manufactured Material," *The Minerals, Metals and Materials Society 2021 Virtual*, Mar. 15–18, 2021.

INTELLECTUAL PROPERTY:

Downey, C.M., I. v. Rooyen, L. Nuñez, "Fabrication of High Entropy Alloys with Additive Manufacturing Utilizing Commercial Alloys and in situ Process Control," Patent application (Dec. 21, 2021).





Three-dimensional finite element model generated by injecting random elliptical pores to represent both pores and cracks in addition to tristructural isotropic particles distributed based on a fitted distribution coming from the x-ray computed tomography scans. (a) space exploration and training data generation. (b) experiment-informed modeling from Advanced Gas Reactor compacts. (c) using Raven and Python to build an Abaqus model that matches the tristructural isotropic particles distribution. (d) resulting distribution to be compared to (b). (e) resulting finite element model to be used in training the machine learning models. (f) reference simulation of the finite element model.

Real-time Non-equilibrium Studies for Nuclear Materials Performance

PROJECT NUMBER:
21A1055-003

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PRINCIPAL INVESTIGATOR:
Fidelma Di Lemma

CO-INVESTIGATORS:
Lingfeng He, INL
Joerg Jinschek, The Ohio State University
Sriram Vijayan, The Ohio State University

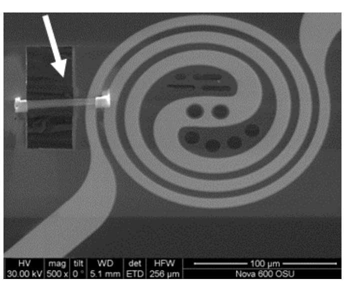
Innovative transmission electron microscope modification leads to faster and cheaper nuclear fuel qualification.

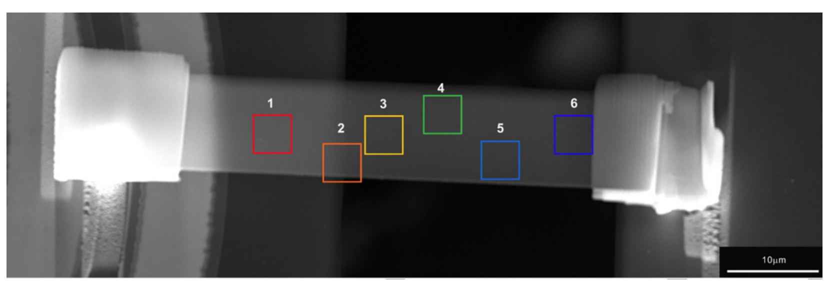
This project developed and tested a new device that can replicate the thermal gradients experienced in sodium-cooled fast reactors in a transmission electron microscope. Such capability is not commercially available. This device can provide insights into thermally driven phenomena that govern the microstructural evolution of nuclear materials and enable evaluation of the microstructure and phase evolution in real time. These studies can reduce the time and cost of experiments associated with testing and qualification of new nuclear materials and fuel. Indeed, the proposed device can reduce irradiation campaign by down-selecting materials based on their thermal behavior before irradiation testing is conducted.

In this project, the feasibility of obtaining a prototypical thermal gradient on the order of 10^4 C/m was confirmed. The temperature was measured via the sublimation of silver nanocubes. Moreover, the desired sample configuration for the experiments was optimized. This included engineering a sublayer to minimize sample interaction with the heating element. Testing of this capability on nuclear fuel, such as uranium-zirconium and uranium-molybdenum alloys, was performed and provided scientific insight into phase and microstructure transition under thermal gradients. Phase segregation varied across the thermal gradient and grain nucleation was observed to start in the hot region of the thermal gradient preferably at grain boundary triple junctions. The setup was also applied to study rare earth behavior in metallic fuel. The experiment highlighted the capability of the device to monitor elemental redistribution. As rare earth elements were found to redistribute from the hot region to the cold region.

TALENT PIPELINE:

- Sriram Vijayan, postdoc at The Ohio State University
- Kaustubh K. Bawane, postdoc at INL, converted to staff





Left: The modified micro electromechanical system heater and the mounted sample. Right: The thermal gradient across the sample is monitored via sublimation of silver nanocubes.

Flash Neutron Radiography at the Transient Reactor Test Facility

PROJECT NUMBER:
20A44-200

TOTAL APPROVED AMOUNT:
$1,046,000 over 3 years

PRINCIPAL INVESTIGATOR:
Aaron Craft

CO-INVESTIGATORS:
Shawn Jensen, INL
William Chuirazzi, INL
Joshua Gess, Oregon State University
Wade Marcum, Oregon State University
Burkhard Schillinger, Technical University Munich
Steven Cool, DMI/Reading Imaging

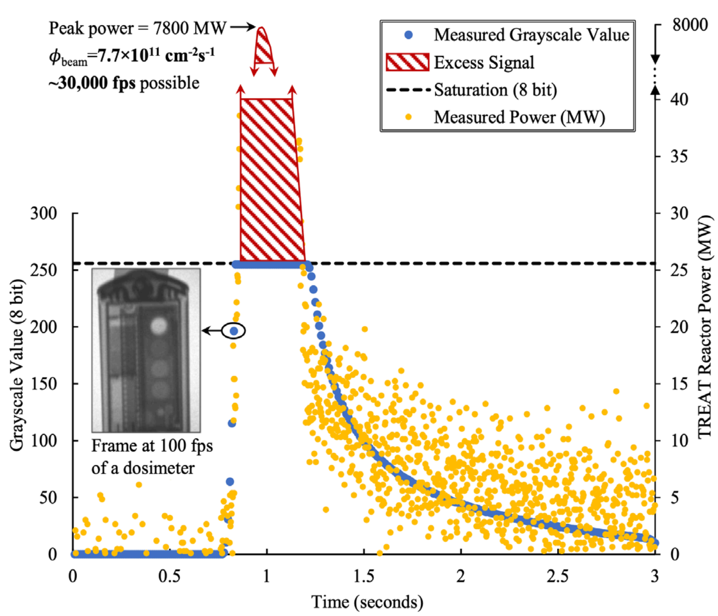
High-speed neutron imaging system enables science that was previously impossible.

Flash neutron radiography can visualize high-speed events that are impossible to see with other techniques. Visualizing bubbly flow at prototypical temperatures and pressures of a pressurized water reactor, for example, is not possible with any other technique. Previous attempts were limited by the available neutron beam intensity. During a power transient, the Transient Reactor Test facility neutron radiography beam is roughly 1000 times brighter than the current brightest neutron imaging beams in the world, representing a shining opportunity to establish unique experimental capabilities at INL.

This project modified the neutron beam so it could be used during a transient and developed a high-speed digital neutron radiography system capable of frame rates faster than 1 millisecond. To further improve the imaging system and more efficiently use the available neutrons, researchers developed neutron imaging scintillator screens using boron-10 that offer higher neutron detection efficiency compared to the most common neutron imaging screens that use lithium-6.

The reactor modifications are complete, operation of the beam during a Transient Reactor Test facility transient is approved, and the flash neutron radiography system is built. The first measurements produced a neutron beam flux of 7.7×10^{11} $cm^{-2}s^{-1}$ during the 7,800 MW peak transient, which is the brightest neutron imaging beamline in the world. This neutron flux would be sufficient for frame rates of ~30,000 fps, far surpassing the current state-of-the-art of ~1000 fps.

This project represents a worldwide collaboration between INL, Reading Imaging, Oregon State University, and Technical University Munich. Several journal articles are published based on this project. This new high-speed neutron radiography capability represents a new state-of-the-art for high-speed neutron radiography that enables science that would previously impossible.



Results from flash neutron radiography of a dosimeter acquired during a transient with a peak power of 7,800 MW. The camera sensitivity was at its maximum setting and the frame rate was 100 fps. The images saturate quickly during the measurement, demonstrating the potential to acquire images at ~30,000 fps with the peak neutron beam flux of 7.7×10^{11} $cm^{-2}s^{-1}$.

TALENT PIPELINE:

- William Chuirazzi, *Russell L. Heath Distinguished Postdoctoral Fellow*, converted to staff
- Sophia Brodish, student at Oregon State University

PUBLICATIONS AND PRESENTATIONS:

Chuirazzi, W., A. Craft, "Measuring thickness-dependent relative light yield and detection efficiency of scintillator screens," *Journal of Imaging* 6(7) (2020) 56.

Chuirazzi, W., A. Craft, B. Schillinger, S. Cool, A. Tengattini, "Boron-based neutron scintillator screens for neutron imaging," *Journal of Imaging* 6(11) (2020) 124.

Schillinger, B., W. Chuirazzi, A. Craft, S. Cool, A. Tengattini, "Performance of borated scintillator screens for high-resolution neutron imaging," *Journal of Radioanalytical and Nuclear Chemistry*, (2022).

Schillinger, B., "New Measurements with High-Resolution Borated Neutron Imaging Screens," *12th International Conference on Methods and Applications of Radioanalytical Chemistry (MARC-12)*, Apr. 3–8, 2022.

Craft, A., "A high flux transient neutron beam for flash neutron radiography of highly dynamic processes at the Transient Reactor Test Facility," *9th International Topical Meeting on Neutron Radiography*, Oct. 2022.

Craft, A., S. Jensen, J. Gess, W. Marcum, L. Warby, S. Brodish, J. Mendoza, W. Chuirazzi, "A high flux transient neutron beam for flash neutron radiography of highly dynamic processes at the Transient Reactor Test Facility," *9th International Topical Meeting on Neutron Radiography*, Dec. 2022.

Modeling Internal Material Melting

PROJECT NUMBER:
21A1055-020

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PRINCIPAL INVESTIGATOR:
Adam Zabriskie

CO-INVESTIGATOR:
Stephen Novascone, INL

Internal melting and solidification are modeled in state-of-the-art nuclear fuel performance simulation code.

To model internal material melting or solidifying, the enthalpy form of the heat conduction governing equation was implemented into BISON and MOOSE conduction module. During a phase change produced from internal nuclear heating or after as the fuel cools, the energy change is not entirely reflected by the change in temperature due to the required heat of fusion energy of the phase change. For heat conduction governing equation models that solve for the temperature field solution, treatment of this behavior can be complicated. The enthalpy form of the heat conduction equation solves for the enthalpy field solution, which continuously changes even during phase change as energy is changed. MOOSE kernels for each term in the governing equation were created. A MOOSE fixed temperature Dirichlet boundary for the enthalpy form was created. Temperature-dependent solidus and liquidus temperature material models for uranium-plutonium-zirconium were added to BISON. All code additions include documentation of the theory and user manual and regression tests for software quality assurance requirements of both MOOSE and BISON. Both standard and automatic differentiation forms were provided for the new code. Unfortunately, an enthalpy form material model converter was not completed, preventing the use of the created code on a BISON assessment.

$$\rho \frac{\partial E}{\partial t} = \nabla \Gamma \nabla E + \nabla^2 S + \dot{q}$$

The enthalpy form of the heat conduction equation where ρ is density, E is a change in enthalpy from a reference state enthalpy, Γ and S are derived material properties from specific heat, thermal conductivity, and heat of fusion, and $\dot{q}$ is a volumetric heat source.

Developing Generalized Methods for Handling Transmutation Radiation Damage

PROJECT NUMBER:
21A1055-022

TOTAL APPROVED AMOUNT:
$131,400 over 1 year

PRINCIPAL INVESTIGATOR:
Micah Gale

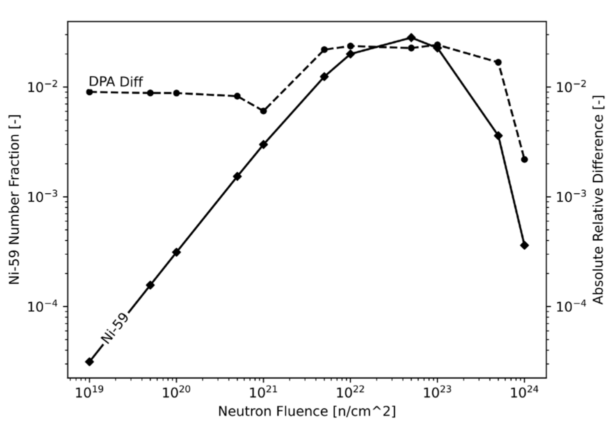
CO-INVESTIGATORS:
Angelica Mata Cruz, INL
Sebastian Schunert, INL

New computational method calculates radiation damage from all isotopes, even those produced during irradiation, in a single efficient calculation.

Accurate models for radiation damage are crucial for predicting material performance in radiation environments. The uncertainty of state-of-the-art radiation damage models is large, contributing to excessive safety margins. A major source of this uncertainty is neglecting the effect that transmutation products have on radiation damage. Transmutation products are new nuclides formed by neutron activation during irradiation. They can contribute to radiation damage by additional neutron capture or decay events. Ignoring the contribution of transmutation products leads to a significant underprediction of the radiation damage (e.g., >10% error in 316 stainless steel). This underprediction is accounted for in part by adding larger safety margins to designs. Prior to this project, the state-of-the-art explicitly accounted for only a single transmutation product, nickel-59, during the radiation damage calculation. All other transmutation products were assumed to not contribute to the radiation damage because there was no established method to systematically track all or a selection of radiation damage contributions of transmutation products during activation. In the case of nickel-59, the past method was to apply a precalculated correlation that cannot be used for any other nuclide and is largely dependent on all nuclear engineers being experts in this niche topic.

We methodically found other transmutation products that cause significant radiation damage, developed a general framework for systematically tracking the radiation damage from these nuclides, and developed a model for radiation damage from radioactive decay. This new framework combines the radiation damage calculation into the transmutation calculation already performed for irradiated structural materials. The key idea of our framework was to introduce radiation damage "pseudo-nuclides" to the list of nuclides used in the transmutation analysis. This allowed radiation damage to be tracked alongside the creation and destruction of transmutation products. This capability was implemented and demonstrated in INL's neutronics code Griffin. This method was able to calculate radiation damage within 2.5% of a published analytical benchmark with the error likely coming from having to reverse engineer the data the benchmark used.

Absolute relative difference in radiation damage measured in displacements per atom between Griffin and the Greenwood analytical benchmark alongside nickel-59 number fraction.



TALENT PIPELINE:

- Olin Calvin, student at University of Idaho
- Daniel Van Wasshenova, student at Idaho State University

PRESENTATION:

Gale, M., O. Calvin, and S. Schunert, "Using Griffin's Transmutation Solver to Calculate Radiation Damage," *International Conference on Physics of Reactors 2022*, Pittsburgh, PA, USA, May 2022.

Multi-principal Element Alloy Fuels for Fast Reactors

PROJECT NUMBER:
21A1057-007

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PRINCIPAL INVESTIGATOR:
Geoffrey Beausoleil II

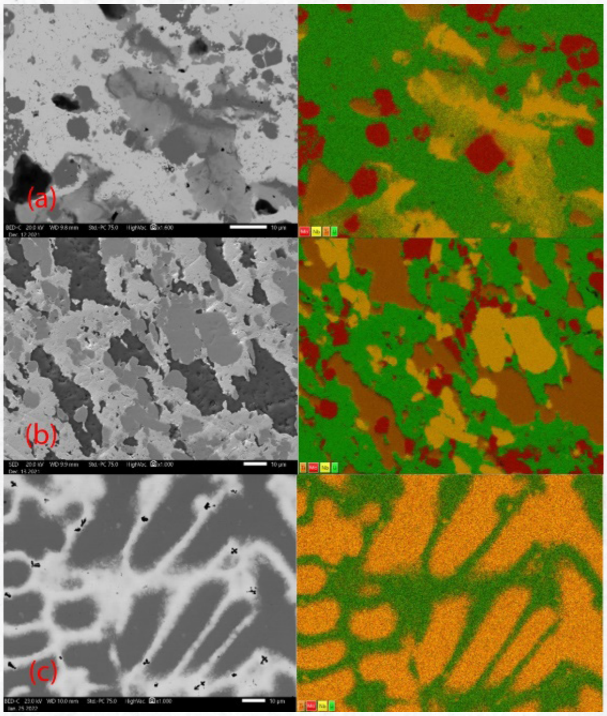
CO-INVESTIGATORS:
Nathan Jerred, INL
James Zillinger, INL
Xiaofei Pu, INL

Actinide bearing multi-principal element alloys that could be used for high temperature reactor fuel were fabricated for the first time.

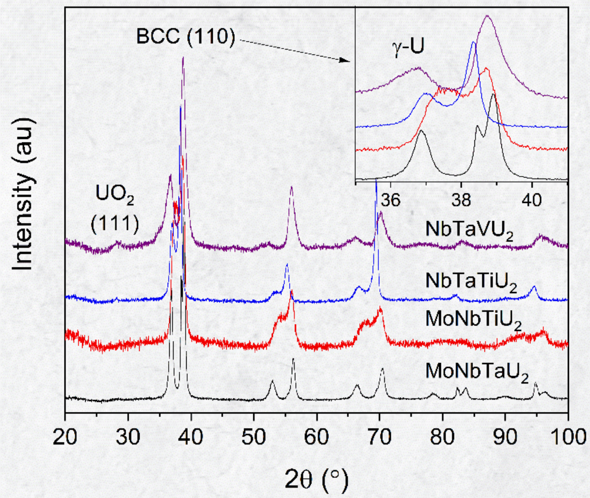
This research explored the fabrication and microstructure of a uranium (U) bearing multi-principal element alloy for use as a high-assay low enriched uranium fuel material. The incentive of these compositions was to create an alloy system that remains single phase body-centered cubic from room temperature through anticipated operation temperatures to improve fuel performance and predictability. Previous research has shown that multi-principal element alloys using chromium, molybdenum (Mo), niobium (Nb), tantalum (Ta), titanium (Ti), vanadium (V), and zirconium can form stable body-centered cubic structures across a large temperature range (25-1000°C). This is the same structure and space group as gamma phase uranium that has shown desirable behavior in previous alloy fuel research. These elements, along with uranium, were assessed through materials property databases, and predictors were used to determine compositions with a high potential of forming a solid solution alloy. These compositions were also analyzed using Monte Carlo n-Particle analysis to determine uranium densities necessary to make the alloy a viable fuel. These two approaches resulted in a down selection of four alloys: $MoNbTaU_2$, $MoNbTiU_2$, $NbTaTiU_2$, and $NbTaVU_2$. These alloys were fabricated using spark plasma sintering of raw elemental powders and through arc melting of raw elemental foils. Characterization of the fabricated alloys included scanning electron microscopy, x-ray diffraction, and energy dispersive x-ray spectroscopy. The results showed that mixing and consolidation through powder methods, ball milling, and spark plasma sintering was inadequate for producing a homogenized alloy with all elements remaining mostly segregated from each other. The arc-melted samples, however, produced a two-phase system with the uranium forming a body-centered cubic matrix phase around a body-centered cubic solution of the other alloying elements. While this work did not yield its intended result, the processing did allow for a uranium rich body-centered cubic phase to form and could lend itself to improved opportunities for future fuel fabrication development.

TALENT PIPELINE:

- Abdullah Weiss, student at Texas A&M University



A comparison of the scanning electron micrographs and energy dispersive x-ray spectroscopy maps of the $MoNbTiU_2$ alloy. (a) High temperature and low pressure spark plasma sintering sample. (b) Low temperature and high pressure spark plasma sintering sample. (c) Arc-melted sample.



X-ray diffraction patterns of the arc-melted samples showing the formation of body-centered cubic phases in the alloys.

Accelerate Multiphysics Solves Using Deep Learning

PROJECT NUMBER:
21A1057-027

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

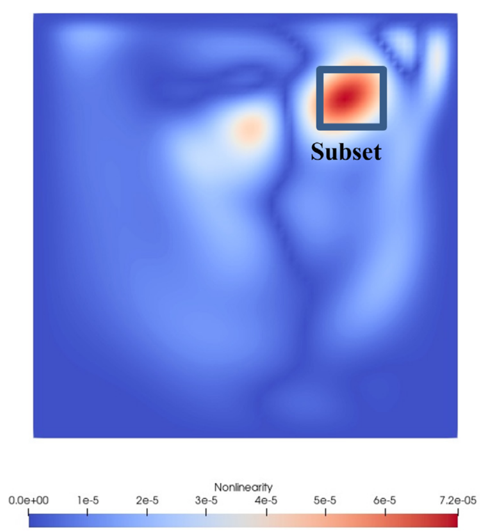
PRINCIPAL INVESTIGATOR:
Fande Kong

CO-INVESTIGATOR:
Alexander Lindsay, INL

Advanced multiphysics solve techniques using modern machine learning benefit research in all computational science domains that depend on nonlinear solution strategies.

Multiphysics simulation is essential in computational science. For decades, multiphysics simulation has been extensively investigated because the phenomena it models are encountered in many applications of interest to DOE and other national agencies. However, solution time significantly limits the applicability of multiphysics simulation. In particular, variation in nonlinearity in multiphysics systems is the leading cause of numerical difficulties in standard simulation algorithms.

This project explored advanced nonlinear solver techniques, including a nonlinear subset accelerator and machine learning techniques. The basic idea of the nonlinear subset accelerator was to form a nonlinear subproblem using the components with high residual values, which correspond to high nonlinearity. The solution of the nonlinear subset accelerator was employed as the initial guess for each nonlinear iteration of the outer solver. We verified the algorithm's effectiveness using a computational fluid dynamics example, a lid-driven cavity. The new algorithm worked for certain problems with small viscosity while the traditional approach did not. In addition, we initially explored the application of deep learning and machine learning to improve the developed nonlinear solver.



Nonlinearity for lid-driven cavity, based on which a nonlinear accelerator was created to accelerate the solver performance.

Coupled Multiphysics Safety Analysis of Molten Salt Reactors Subjected to Earthquakes

PROJECT NUMBER:
21A1057-029

TOTAL APPROVED AMOUNT:
$143,000 over 2 years

PRINCIPAL INVESTIGATOR:
Guillaume Giudicelli

CO-INVESTIGATORS:
Abdalla Abou-Jaoude, INL
Chandu Bolisetti, INL
Kyung Tae Kim, INL
Mauricio Tano, INL
Paolo Balestra, INL

Multiphysics coupling between structural dynamics, neutronics, and thermal fluids transport reduces development time and cost of new molten salt reactors.

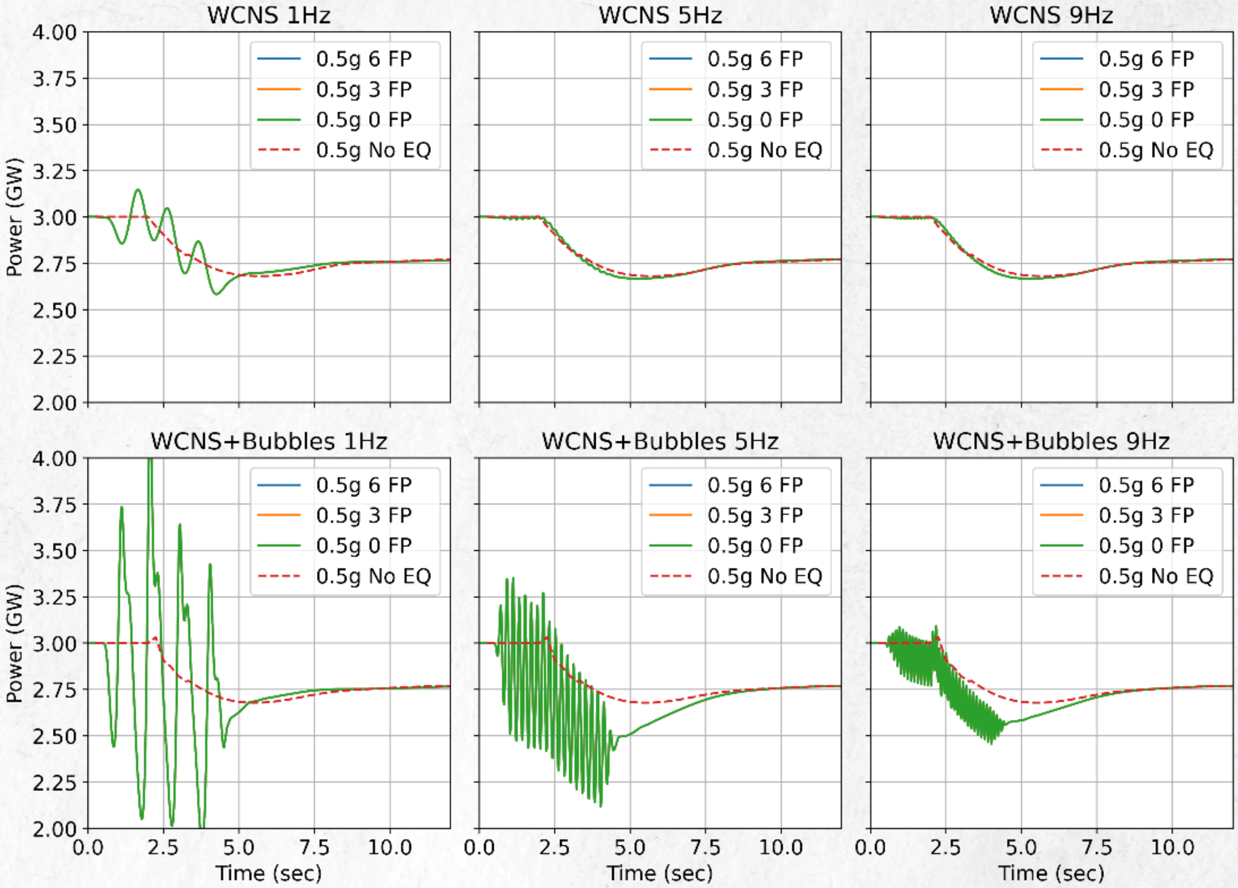
This research developed a proof-of-concept demonstration of a high fidelity coupled multiphysics simulation of a molten salt reactor shaking during an earthquake. The demonstration leveraged MOOSE and used three MOOSE-based applications: (1) Mastodon, to simulate structural dynamics and mechanics from seismic shaking and acoustic wave propagation in the molten salt, (2) Pronghorn, for reactor thermal hydraulics, and (3) Griffin, for neutronic feedback. Molten salt reactors are being pursued by a range of reactor developers but have little precedence apart from an experimental facility built in the 1950s. Their safety and licensing cases can be significantly strengthened by a thorough evaluation of their behavior during earthquake-based transients, ultimately aiding their deployment. Furthermore, significant cost savings can be envisaged from reducing conservatism because earthquake-based considerations are key cost drivers.

The project started with independent, uncoupled molten salt reactor simulations using Mastodon for seismic analysis and Griffin plus Pronghorn for neutronics and thermal hydraulics. The research then progressed by gradually increasing the level of coupling by transitioning from loose coupling to a tightly coupled simulation using the MOOSE 'MultiApp' system where the seismic accelerations in the molten salt are transferred from Mastodon to Pronghorn plus Griffin, which in turn, provided the temperature and density fields to Mastodon at each timestep. This first-of-a-kind coupled multiphysics simulation analyzed a wide range of complex seismic transients and demonstrated a large impact of lower frequency vibration on the power production of the core in an uncontrolled transient. It provided deep novel insights into the level of coupling needed between the seismic analysis and reactor dynamics to accurately simulate earthquake transients. Unrelaxed fixed-point iteration between each physics satisfactorily converged, allowing for an optimization of the coupling scheme based only on the time stepping requirements of each physics.

PRESENTATIONS:

Bolisetti, C., K. T. Kim, G. L. Giudicelli, P. Balestra, S. L. N. Dhulipala, A. Abou-Jaoude, "Coupled Multiphysics Simulations of Molten Salt Reactors Subjected to Earthquakes," *2021 American Nuclear Society meeting*, Washington DC, USA, Nov. 30–Dec. 3.

Bolisetti, C., G. L. Giudicelli, P. Balestra, S. L. N. Dhulipala, M. E. Tano, A. Abou-Jaoude, "Multiphysics Coupled Seismic Safety Analysis of Molten Salt Reactors," *2022 American Nuclear Society meeting*, Phoenix, AZ, USA, Nov. 13–17.



Preliminary results showing the influence of modeling decisions on coupled multiphysics analysis of an earthquake in a molten salt reactor.

Oxidation of Cerium Nitride under Irradiation

PROJECT NUMBER:
21P1062-019

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PRINCIPAL INVESTIGATOR:
Lingfeng He

CO-INVESTIGATORS:
Chao Jiang, INL
Yi Xie, Purdue University

COLLABORATOR:
Texas A&M University

Cerium nitride, a non-radioactive surrogate for actinide nitrides, oxidizes at very low oxygen partial pressures.

Cerium nitride, a uranium nitride fuel surrogate material, was successfully sintered within tens of seconds using a pressureless fast heating technique. The as-sintered pellets have a relative density of 96% with uniform grain size of 10 – 20 µm. Cerium nitride remained stoichiometrically stable during the fast sintering. However, cerium nitride ceramics are very sensitive to air. Transmission electron microscopy characterization of lamellas prepared from the as-sintered cerium nitride pellets shows that the lamella was fully oxidized and became cerium dioxide. Ab initio density functional theory was used to calculate the thermodynamic driving forces for the oxidation of metal nitrides. The calculated large values indicate that the oxidation of metal nitrides is highly favored and can occur even at very low oxygen partial pressures.

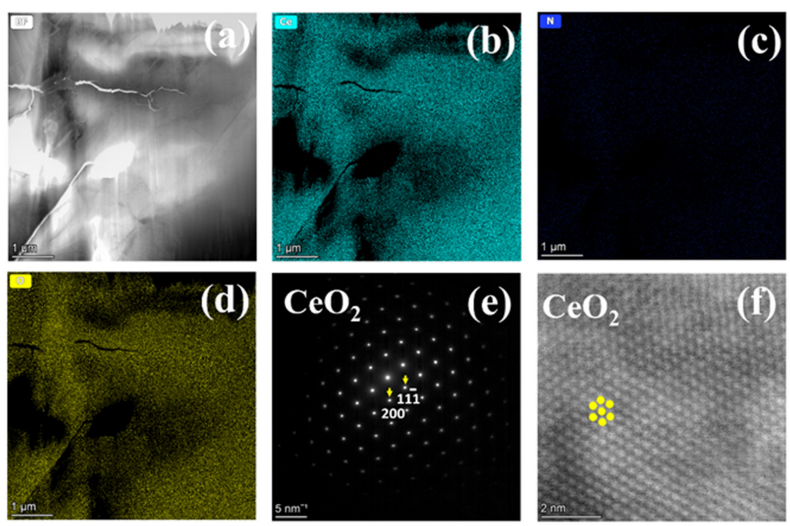
TALENT PIPELINE:

- Logan Joyce, student at Purdue University

PRESENTATIONS:

Joyce, L., Y. Xie, "Fabrication of CeN (UN fuel surrogate) using pressureless fast heating system," *2022 ANS Annual Meeting*, Jun. 12–16, 2022.

He, L., K. K. Bawane, T. Yao, P. Xiu, C. Jiang, M. Khafizov, M. Jin, Y. Xie, L. Shao, "Microstructural Evolution in Ceramic Nuclear Fuels and their Surrogates under Irradiation," *Materials Science & Technology technical meeting (MS&T 22)*, Oct. 2022 (Invited talk).



Characterization of cerium nitride oxidation. (a) Scanning transmission electron microscopy (STEM) image, (b)-(d) energy dispersive x-ray spectroscopy, (e) selected area electron diffraction pattern, and (f) atomic-resolution STEM image of a lift-out lamella from an as-sintered cerium nitride pellet showing that cerium nitride was fully oxidized to cerium dioxide (CeO_2).

Development and Demonstration of Spatial Correlation Techniques among Diverse Data Sets

PROJECT NUMBER:
21P1056-001

TOTAL APPROVED AMOUNT:
$200,000 over 2 years

PRINCIPAL INVESTIGATOR:
William Chuirazzi
Russell L. Heath Distinguished Postdoctoral Fellow

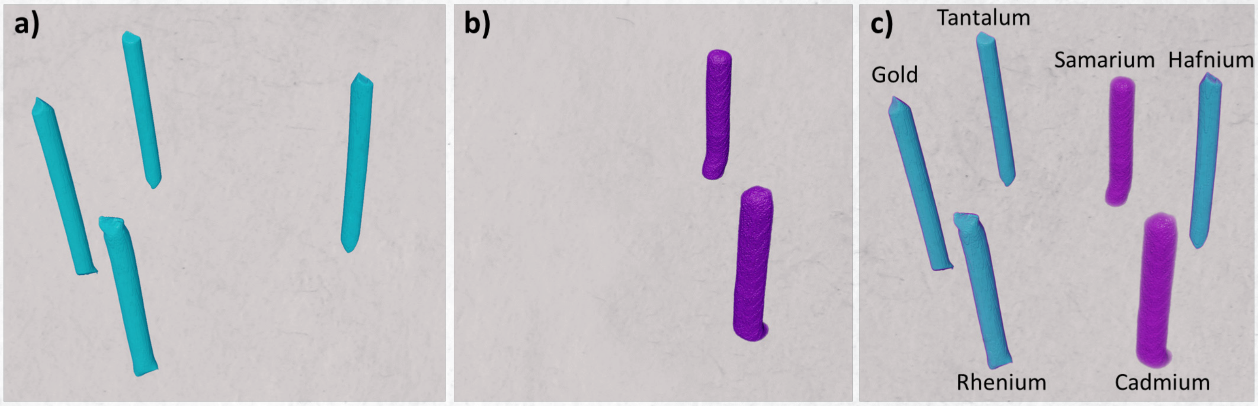
Combining three-dimensional x-ray and neutron imaging data accelerates nuclear energy material development and advances the fundamental understanding of material properties.

State-of-the-art characterization capabilities are needed to improve our knowledge of irradiated fuels and materials. One of the next steps in advanced data analysis is the spatial connection of advanced instrumentation to combine or fuse data together. This provides researchers with a more robust and effective picture of material and may increase the speed and efficiency of post-irradiation examination. This research focused on developing image registration methodologies to practically implement the spatial fusion of data sets. Combining images obtained with different techniques into a single data set allows researchers to better identify material and elemental composition of different sample regions, as well as potentially improve contrast, resolution, or both of certain features.

Before these capabilities can be routinely available to researchers, the workflow for geometrically orienting samples in different instruments and then performing image registration and data fusion must be designed and implemented. During this project, a sample containing different wires relevant to nuclear applications was fabricated, and x-ray tomography and neutron tomography were performed. The sample consisted of samarium, gold, rhenium, cadmium, hafnium, and tantalum, which are materials with nuclear applications and interact with neutrons and x-rays differently, emphasizing how the techniques complement one another.

Simulations and calculations were conducted to determine how each material would perform when imaged, and these results were compared with the experimental images. These data sets were then spatially registered and fused together to create a single data set containing more information than either single data set. This process was then applied to a surrogate molten salt waste form.

This project demonstrated the workflow and proof-of-principle of performing image registration and fusion with x-ray and neutron tomography, enabling multi-modal imaging for INL-relevant samples. The expertise developed from this project allows correlative multi-modal imaging to support future programmatic interests.



Three-dimensional renderings of a sample containing wires of materials relevant to the nuclear industry constructed from a) x-ray tomography data and b) neutron tomography data. c) The reconstructions are spatially registered, so they accurately show the locations of all wires relative to one another. By comparing relative intensities in both data sets, the wire materials can also be identified.

TALENT PIPELINE:

- William Chuirazzi, *Russell Heath Distinguished Postdoctoral Fellow*, converted to staff

PUBLICATIONS AND PRESENTATIONS:

Chuirazzi, W., J. Kane, A. Craft, J. Schulthess, "Image fusion for neutron tomography of nuclear fuel." *Journal of Radioanalytical and Nuclear Chemistry* (2022) 1–7.

Chuirazzi, W., J. Kane, A. Craft, and J. Schulthess, "Image Fusion for Neutron Imaging Applications." *International Conference on Methods and Applications of Radioanalytical Chemistry*, Apr. 2022.

Chuirazzi, W., J. Kane, A. Craft, "Correlative Imaging with Neutron and X-ray Tomography." *International Topical Meeting on Neutron Radiography*, Oct. 2022.

Variationally Consistent Contact Dynamics for General Nuclear Fuel Modeling

PROJECT NUMBER:
21P1062-016

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

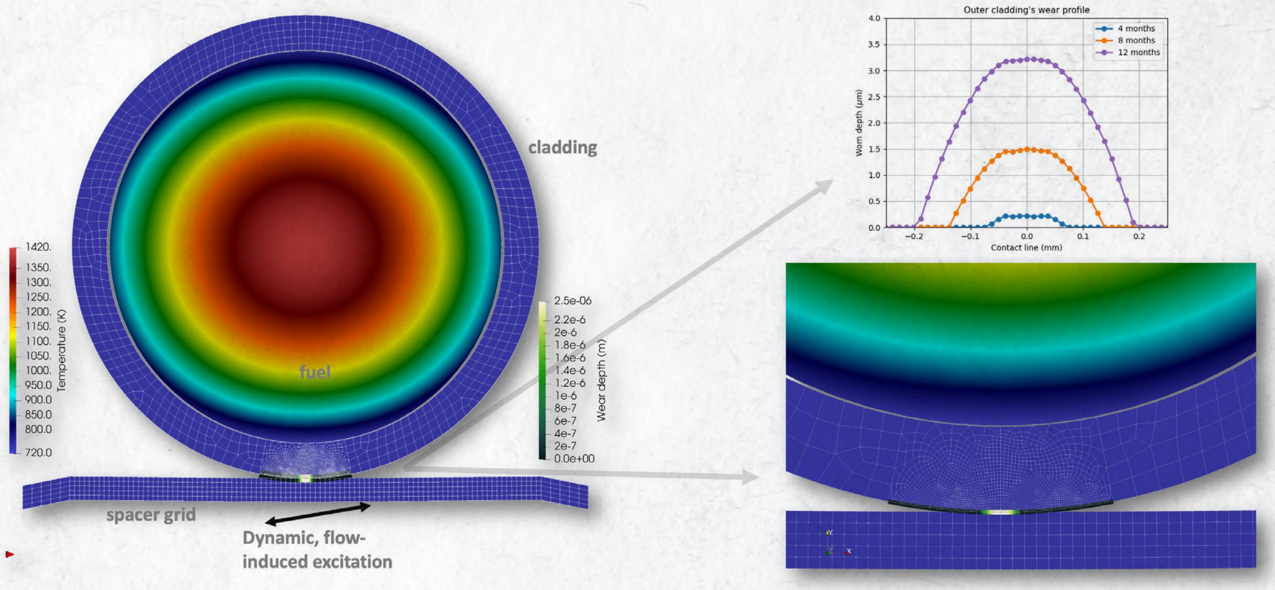
PRINCIPAL INVESTIGATOR:
Antonio Martin Recuero

CO-INVESTIGATOR:
Alexander Lindsay, INL

Novel modeling methodology increases understanding of mechanical contact dynamics and predicts grid-to-rod fretting wear.

Mechanical contact is a pervasive physical phenomenon that entails modeling and numerical challenges. Traditional approaches employ discretized points and surfaces to enforce contact constraints (i.e., "strong" enforcement). More advanced and recently developed approaches, such as the mortar finite element, enforce those constraints on segments in dynamically-computed lower dimensional domains. This work developed the stabilization of mortar contact constraints for dynamic applications—applications in which inertia and vibrations or impacts are of relevance—in MOOSE. A comparative study was completed between a mortar approach and a point-to-surface approach to contact dynamics, which stressed the numerical convergence and accuracy gains obtained by the extension of mortar constraints to dynamic problems.

In the context of mortar-based contact dynamics formulations, small wear depths in the constraint equations were considered, which enables wear in nuclear reactor systems to be studied, such as grid-to-rod fretting. Existing creep, fission gas release, burnup, swelling, thermal expansion, and fuel relocation models in the nuclear fuel performance code BISON were used to run fuel-cladding-spacer grid setups in which dynamic excitations lead to wear on the cladding's outer surface. This methodology allows for the study of grid-to-rod fretting wear by employing a consistent formulation in contact dynamics natively in a nuclear fuel performance code. Various geometries of spacer grid and the influence of fuel-cladding contact were considered. Grid-to-rod fretting wear results were consistent with experimental results and allowed to gain insight into single fuel rod's configurations natively in a MOOSE application. Particularly, the additional stiffness provided by fuel-cladding contact was shown to generate higher normal contact pressure values that are conducive to increased wear generation. Using conformal geometry for spacer grids reduced and spatially homogenized overall wear throughout the contact interface. These findings are consistent with laboratory experiments reported in the literature.



Repeated dynamic excitation causes wear on the cladding outer surface. Wear profile and depth are heavily influenced by the shape of contacting surfaces and severity of contact, which may be determined by internal fuel-cladding mechanical interaction.

PRESENTATION AND PUBLICATION:

Recuero, A., A. Lindsay, "On practical aspects of variational consistency in contact dynamics," *American Society of Mechanical Engineers (ASME) IDETC/CIE Conference*, St. Louis, MO, Aug. 14–17, 2022.

AWARD:

Best paper award at ASME IDETC/CIE 2022 Multibody Systems and Nonlinear Dynamics, St. Louis, MO, United States of America.

Understanding Non-equilibrium, Nanoscale Defects in Bulk Metals

PROJECT NUMBER:
21P1064-008

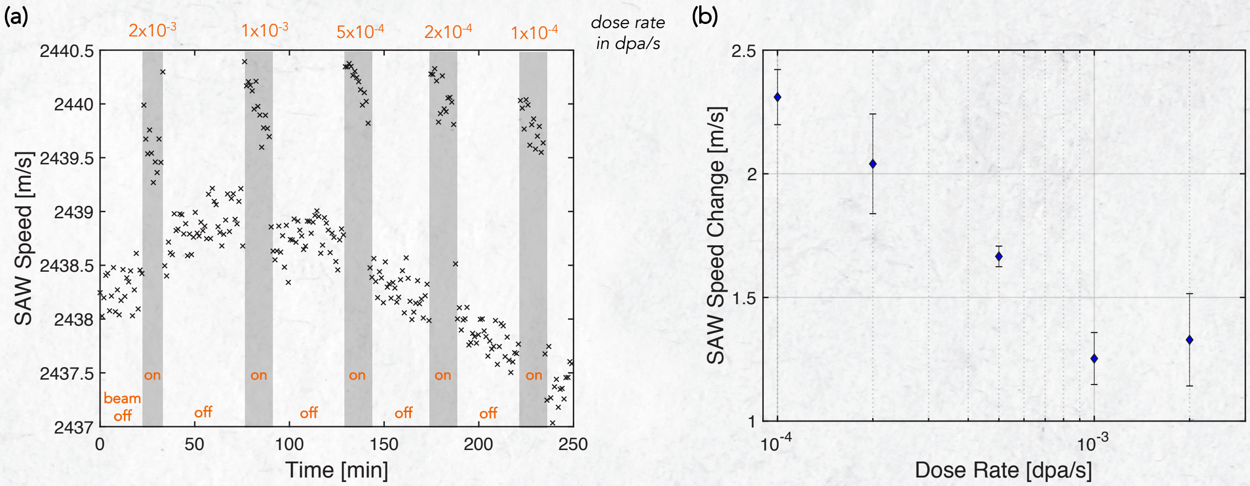
TOTAL APPROVED AMOUNT:
$88,500 over 1 year

PRINCIPAL INVESTIGATOR:
Cody Dennett

Innovative experimental method quantifies material damage during irradiation.

Radiation effects in materials are a concern for any structure exposed to ionizing radiation. Nuclear power applications, accelerators, space nuclear applications, and isotope production facilities all share a common concern of materials property changes during exposure to radiation. Neutron, ion, and electron radiation can transfer energy to atoms in a crystal lattice. If the transferred energy exceeds the displacement energy of the atom, an atomic displacement is produced, which leads to vacancies and interstitials. The quantity of vacancies and interstitials depends on the type of radiation, incoming energy, and the host material. Following the initial displacement, only a fraction of the generated vacancies and interstitials remain and build to a non-equilibrium steady-state concentration of point defects. After the irradiation, when defect generation ceases, the vacancy and interstitial content reduces toward thermodynamic equilibrium. However, it is this initial non-equilibrium defect concentration that is responsible for the formation of larger defects, such as voids or dislocation loops, which can significantly alter material properties. Therefore, understanding and quantifying the non-equilibrium defects in a material is key to predicting long-term property changes. Quantifying non-equilibrium vacancies during irradiation using methods such as in situ positron annihilation spectroscopy can provide partial validation of rate theory models. However, full validation for both vacancies and interstitials must rely on different methods due to the nature of positron-electron recombination.

Researchers used pulsed ion beam irradiation to generate transient elevated populations of nanoscale defects in pure single crystal nickel while in situ laser metrology continuously captured the resulting change in elastic moduli, which are sensitive to both vacancies and interstitials. An initial demonstration of this measurement was completed, but a detailed exploration of defect generation, temperature, ion beam energy, and host material is required. Such data can be used to validate this methodology as a companion for positron annihilation measurements of vacancy concentration and as a benchmark for defect accumulation and evolution codes such as cluster dynamics. These experiments offer a singular lens into the type of transient defect generation and evolution that plays a deterministic role in nuclear system performance.



(a) Time-dependent record of surface acoustic wave (SAW) velocity in pure nickel subject to impulse irradiations of 31 MeV nickel ions at 500°C using in situ time-dependent transient grating spectroscopy. (b) Impulse velocity step-changes resulting from irradiations at different dose rates. Further evolution beyond the initial interstitial-dominated stiffening response is clearly visible in the short "beam on" periods.

PUBLICATIONS:

Hirst, C. A. and C. A. Dennett, "Toward Quantitative Inference of Nanoscale Defects in Irradiated Metals and Alloys," *Frontiers in Materials* 9 (2022) 888356.

Analysis of Mars Perseverance Multi-mission Radioisotopic Thermoelectric Generator Neutron/Gamma Spectrum for use on Dragonfly and Nuclear Thermal Propulsion Materials Modeling

PROJECT NUMBER:
22P1073-001

TOTAL APPROVED AMOUNT:
$275,000 over 1 year

PRINCIPAL INVESTIGATOR:
Stephen Johnson

CO-INVESTIGATORS:
Stephen Herring, Center for Space Nuclear Research
Brad Kirkwood, Consultant

COLLABORATOR:
NASA Marshall Space Flight Center

Researchers enable faster and safer exploration of the outer solar system by proving the feasibility of nuclear propulsion and electricity generation.

There are two primary needs for nuclear power in space: propulsion and electrical power. This project included five integrated studies for using a reactor for nuclear thermal propulsion for short burns (about 30 minutes) at high power (~500 MWth) and lower power use (about 700 kWe) for about 400 days between the burns. For nuclear thermal propulsion, hydrogen flows first through the structural cooling channels where the temperature increases from 20 K to about 800 K followed by flow through the reactor fuel itself, where the temperature increases from 800 K to about 2800 K. During the low power electrical generating phase, the structural cooling channels carry a helium and xenon mixture in a closed Brayton cycle to produce about 200 kWe.

Specific research tasks during the project included:

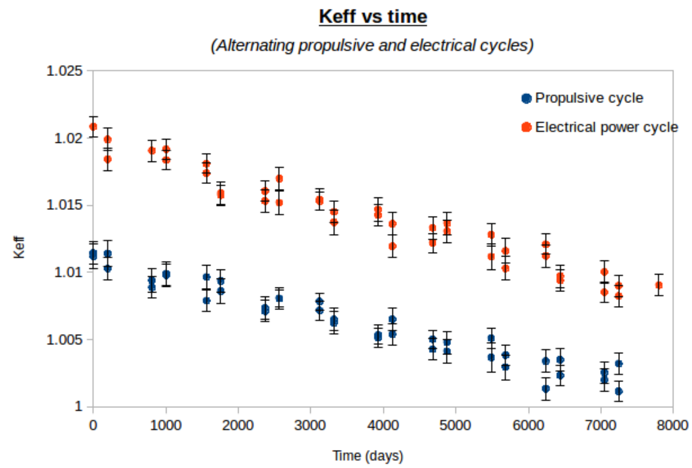
- Describing the Bimodal Epithermal Astro-nuclear Reactor concept
- Completing a thermal structural analysis of flow tube and fuel used in the nuclear thermal propulsion reactor
- Determining the hydrogen diffusion in the flow tube of nuclear thermal propulsion system
- Calculating materials thermophysical properties of uranium zirconium carbide ($U_{0.7}Zr_{0.3}C$) using density functional theory analyses
- Studying the tungsten-184 enrichment to avoid the high neutron absorption of tungsten-186

The Monte Carlo burnup analysis of the bimodal reactor—alternating propulsive and electrical burns—indicates that reactor could power five missions from Earth to Mars and return, where the hydrogen propellant would be replenished on each return to low Earth orbit. Upon the completion of the five missions, the control drums would be removed and replaced by control rods in the same locations. The modified reactor would be emplaced in the lunar or Martian regolith and would produce 700 kWe for about 25 years. Each mission includes:

- 30-minute trans-Mars injection
- 197-day Earth to Mars transfer
- 23-minute Mars orbit insertion
- 555-day Mars parking orbit
- 20-minute trans-Earth injection
- 198-day Mars to Earth transfer
- 15-minute Earth orbit insertion
- 610-day Earth parking orbit

TALENT PIPELINE:

- Zyed Ansary, student at the University of Denver
- Daniel Black, student at Brigham Young University–Idaho
- Aanchal Gupta, student at the University of Illinois Urbana–Champaign
- Kasturi Khatun, student at the University of Southern California
- Kean Martinic, student at Idaho State University
- Manikandan Pandiyan, student at the University of Michigan
- Arnold Pradhan, student at the University of Idaho
- Berenice Sosa Aispuro, student at Idaho State University
- Daniel Watson, student at Texas A&M University
- Teyen Widdicombe, student at the University of Idaho



Propulsion and electricity for five mars missions.

INTEGRATED FUEL CYCLE SOLUTIONS

INL's extensive facilities and expertise in nuclear fuels and materials creates vital integrated fuel cycle solutions to sustain the current reactor fleet, support the demonstration and deployment of new advanced reactors, and facilitate the management and disposition of existing and future radiological waste materials. By exploring fuel cycle technologies for advanced reactor demonstration and deployment, INL focuses on a fuel cycle with inherent process transparency, reducing proliferation risk and including safeguards and security by design. INL utilizes its core capabilities to advance the availability of special nuclear material and strategic isotopes, manage radiological waste materials and used nuclear fuel, reduce proliferation risk, and develop research, development, and deployment test beds.


V300

Advanced Computational Modeling and Experimental Determination of the Thermophysical Properties of Molten Salt Systems Applicable to Molten Salt Reactor Design

PROJECT NUMBER:
20A44-041

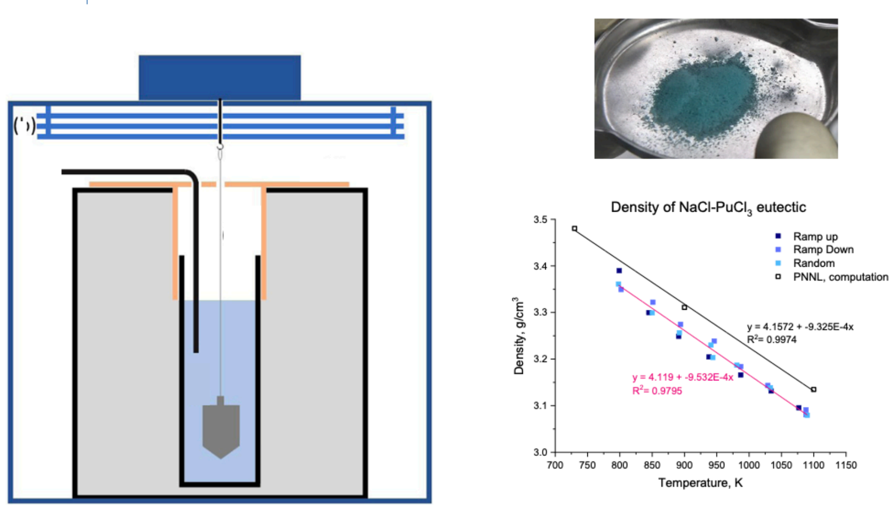
TOTAL APPROVED AMOUNT:
$1,349,000 over 3 years

PRINCIPAL INVESTIGATOR:
Ruchi Gakhar

CO-INVESTIGATORS:
Toni Karlsson, INL
Ben Beeler, North Carolina State University

Cultivating synergies between experiment and modeling expedites thermophysical property measurements on salt systems essential for molten salt reactor advancement.

The fundamental chemistry and properties of molten salts have been the subject of research for decades. Over the past few years, there has been an unprecedented surge in interest for these systems given their apparent advantage in areas related to clean and sustainable energy harvesting and transfer. Despite such focused efforts on molten salt chemistry and properties, the scientific community and molten salt reactor developers still lack understanding of and access to reputable literature data on the thermophysical properties such as density, melting point, viscosity, thermal conductivity, and contact angle of molten salts and how they vary with temperature. Therefore, it is necessary to develop a database of thermophysical properties of multi-component coolant and fuel salt systems. Leveraging INL's unique existing molten salt knowledge base and actinide handling capabilities, the team built an infrastructure to determine fundamental properties of molten salts—a capability unique to INL—specifically for actinide loaded salt systems. Through specialized experimentation, the team obtained high fidelity experimental data on the thermophysical properties for chloride- and fluoride-based molten salts. In addition, ab initio molecular dynamics based predictive computational models were developed and validated against experimental measurements. This allowed the team to step outside of experimental bounds and expand the testing regime to the temperature and compositional ranges difficult to explore experimentally. Another unique aspect of this work is the first ever melt temperature analysis, density measurements, and x-ray diffraction characterization of the sodium chloride plutonium chloride eutectic system, which is applicable to the Molten Chloride Fast Reactor design.



(left) Schematic for Archimedes principle-based molten salt density measurement setup; (right top) Sodium chloride plutonium chloride ($NaCl$-$PuCl_3$) eutectic salt-mixture; (right bottom) density of $NaCl$-$PuCl_3$ eutectic system as a function of temperature.

TALENT PIPELINE:

- Michael Woods, postdoc at INL, converted to staff
- Carl Karlsson, postdoc at INL, converted to staff
- Maria del Rocio Laguna, *Glenn T. Seaborg Distinguished Postdoctoral Fellow*
- Yuzhou Wang, postdoc at INL, converted to staff
- Kai Duemmler, student at North Carolina State University
- Christopher Wolfe, intern at INL

PUBLICATIONS AND PRESENTATIONS:

Duemmler, K., Y. Lin, M. Woods, T. Karlsson, R. Gakhar, B. Beeler, "Evaluation of thermophysical properties of the LiCl-KCl system via ab initio and experimental methods," *Journal of Nuclear Materials* 559 (2022) 153414.

Andersson, D., B. Beeler, "Ab initio molecular dynamics (AIMD) simulations of NaCl, UCl_3 and NaCl-UCl_3 molten salts," *Journal of Nuclear Materials* 568 (2022) 153836.

Duemmler, K., M. Woods, T. Karlsson, R. Gakhar, B. Beeler, "An ab initio molecular dynamics investigation of the thermophysical properties of molten NaCl-$MgCl_2$," *Journal of Nuclear Materials* 570 (2022) 153916.

Duemmler, K., Y. Lin, M. Woods, T. Karlsson, R. Gakhar, and B. Beeler, "Ab initio molecular dynamics study of transport properties in high temperature LiCl-KCl," *American Nuclear Society Winter Conference 2021*.

Duemmler, K., Y. Lin, M. Woods, T. Karlsson, R. Gakhar, and B. Beeler, "Ab initio molecular dynamics study of thermophysical and transport properties for high temperature LiCl-KCl and NaCl-$MgCl_2$ system," *2022 The Minerals, Metals, and Materials Society Annual Meeting & Exhibition, Materials and Chemistry for Molten Salt Systems Symposium* 2022.

Woods, M., T. Karlsson, R. Gakhar, "Wetting properties of molten salts at material interface for reactor applications," *2022 The Minerals, Metals, and Materials Society Annual Meeting & Exhibition, Materials and Chemistry for Molten Salt Systems Symposium* 2022.

Understanding Salt Chemistry and Corrosion Control of Chloride Salts for Molten Salt Reactors

PROJECT NUMBER:
20A44-126

TOTAL APPROVED AMOUNT:
$1,532,900 over 3 years

PRINCIPAL INVESTIGATOR:
Guoping Cao

CO-INVESTIGATORS:
Jianguo Yu, INL
Junhua Jiang, INL
Ruchi Gakhar, INL
Yaqi Wang, INL
Haiyan Zhao, University of Idaho

Understanding fundamental salt chemistry enables corrosion resistant materials development for molten salt reactors.

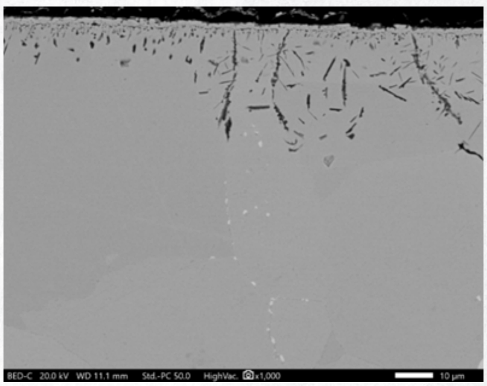
Chloride salts are candidate fuel and coolant for some molten salt reactor concepts as they offer potential for sustained operation in a fast neutron spectrum with minimal salt processing, passive safety, and a high power density. However, molten chlorides present significant corrosion challenges to structural materials. Currently, the corrosion data for relevant metal alloys in chloride salts are very limited, presenting technical risks for reactor design and operation.

This project provided critical baseline data to understand the salt chemistry and corrosion control of two chloride salts—sodium chloride (NaCl)-uranium chloride (UCl_3) and NaCl-magnesium chloride ($MgCl_2$)—most relevant to chloride molten salt reactors. From systematic experimentation and multiphysics based density functional theory modeling, the following major important findings were made on the corrosion of candidate materials for molten salt reactors in chloride salts, salt redox potential control, and guidelines for developing corrosion resistant alloys for chloride molten salt reactors for long-term operation.

- The correct testing methodology for evaluating corrosion of materials is very important. Galvanic corrosion and dissimilar materials effects during corrosion testing can significantly accelerate the corrosion of alloys in chloride salts.
- Sparging a gas mixture of argon and hydrogen into molten chloride salt could dramatically decrease the corrosion of materials in chloride salts.
- The effect of fission product cesium iodide on 316H stainless steel in NaCl-$MgCl_2$ was insignificant.
- After systematic testing of many different iron-based and nickel-based alloys in NaCl-$MgCl_2$ salt under identical testing conditions, it was found that Hastelloy C-276 exhibited outstanding corrosion resistance with a weight loss lower than that of pure nickel. Hastelloy C-22 also exhibited good corrosion resistance.
- Analysis of the systematic corrosion tests on many different alloys could provide fundamental guidelines for developing alloys for chloride molten salt reactors that would exhibit good corrosion resistance and mechanical properties for long-term operations.
- First principles density functional theory multiphysics modeling found tremendous disproportionate behavior and chemical stability difference of UCl_3 and uranium fluoride (UF_3). The modeling shows that UCl_3 is much more stable than UF_3, suggesting different requirements for chloride- and fluoride-fueled molten salt reactors.

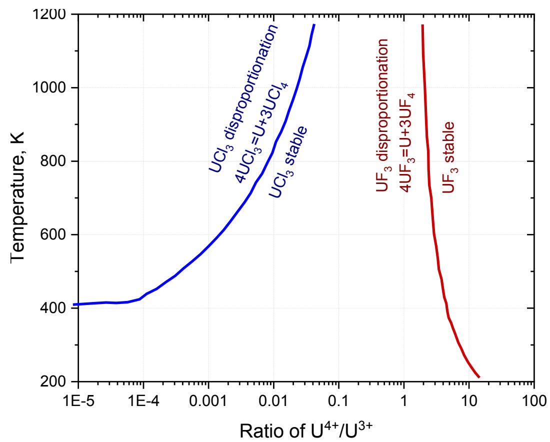
TALENT PIPELINE:

- Drew Glenna, student at University of Idaho
- Trishelle Copeland-Johnson, *Glenn T. Seaborg Distinguished Postdoctoral Fellow*, converted to staff
- Fei Teng, researcher at INL



PRESENTATIONS:

- Glenna, D. M., M. Lawson, H. Zhao, "Corrosion Chemistry and Control in Molten NaCl-$MgCl_2$ Salts," *ANS Transactions* 125(1) (2021) 131-134.
- Copeland-Johnson, T. M., D. J. Murray, G. Cao, "Correlated characterization of Ni-based superalloys corroded in uranium-containing molten salt systems," *MS&T22: Materials Science & Technology: Tackling Structural Materials Challenges for Advanced Nuclear Reactors* 2022.
- Copeland-Johnson, T. M., M. E. Woods, R. Gakhar, D. J. Murray, G. Cao, L. He, "Multi-Modal Characterization of Interfacial Corrosion of Ni-based Alloys in Chloride-based Molten Salts," *The Minerals, Metals, and Materials Society 2022 Symposium on Materials and Chemistry for Molten Salt Systems* 2022.



(left) Corrosion of Alloy 617 in NaCl-$MgCl_2$ salt at 700°C for 1000hrs, showing the aluminum as an oxygen getter which forms alumina, mitigating the corrosion of Alloy 617. (right) Density functional theory modeling result of UCl_3 and UF_3 showing the tremendous disproportionation and chemical stability difference of UCl_3 and UF_3, suggesting the different requirements for molten salt reactors using chloride and fluoride salts.

Development of a Simple Single Step Ultra Chemical Purification of Neptunium

PROJECT NUMBER:
20P1048-005

TOTAL APPROVED AMOUNT:
$351,000 over 3 years

PRINCIPAL INVESTIGATOR:
Thibaut Lecrivain
Glenn T. Seaborg Distinguished Postdoctoral Fellow

CO-INVESTIGATORS:
Kevin Carney, INL
Travis Grimes, INL

COLLABORATOR:
Florida International University

Capillary electrophoresis coupled with advanced complexation chemistry improves lanthanide element separation.

Various methods of separation of lanthanides have been developed because these elements are vital to ongoing technological and energy advancement. The separation of a pure metal, cleaned of other metals, usually requires complex chemistry and a laborious process. To optimize that process, this research coupled capillary electrophoresis to the octadentate siderophore derivative N,N′-(butane-1,4-diyl)bis(1-hydroxy-N-(3-(1-hydroxy-6-oxo-1,6-dihydropyridine-2-carboxamido)propyl)-6-oxo-1,6-dihydropyridine-2-carboxamide), abbreviated 343HOPO, as separation electrolyte. In this project, 343HOPO was synthesized, purified, and characterized for purity.

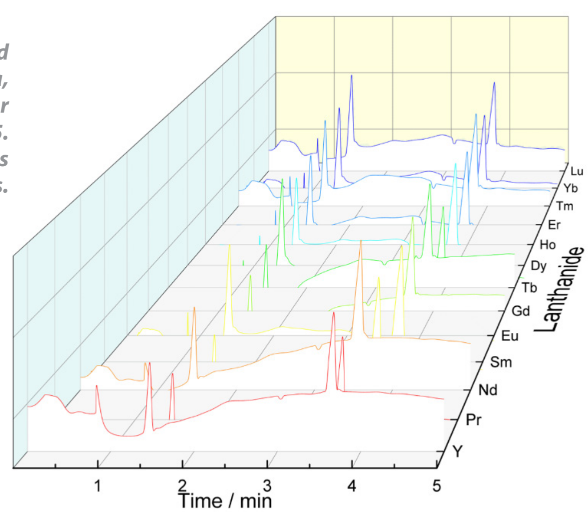
To track separation in the electric field, capillary electrophoresis uses light absorption and negative light absorption methods. In this work, the negative light absorption method could be used because the electrolyte used would have interfered with the electrolyte of interest. Also, classic absorption could not be used because most of the lanthanides do not absorb light in the accessible wavelengths. To overcome these limitations, a contactless conductivity detector from Sensor Technologies was installed, coupled to the capillary electrophoresis, and calibrated. The lower detection limits of the capillary electrophoresis-contactless conductivity detector were determined, and base diffusion/separation in absence of complexing electrolyte was measured. When the complexing electrolyte was used in the system, the separation of each lanthanide, especially late vs. early separation, was significantly enhanced. The observation of this increased separation factor between lanthanides using a capillary electrophoresis system coupled with a complexation agent is considered a major accomplishment and successful demonstration of the method for automating a challenging separation.

TALENT PIPELINE:

- Thibaut Lecrivain, *Glenn T. Seaborg Distinguished Postdoctoral Fellow*

PRESENTATION:

- Lecrivain, T., "Thermodynamic of separation chemistry of f-elements," *American Chemical Society, Nuclear Chemistry Symposium,* 2022.



Electropherogram of selected lanthanides in nitrate media, background electrolyte 5 millimolar sodium nitrate, pH = 4.5. The peaks between 3 and 4 minutes are the lanthanides.

Moving Molten Zone Refining Process Development for Innovative Fuel Cycle Solutions

PROJECT NUMBER:
21A1050-049

TOTAL APPROVED AMOUNT:
$743,000 over 2 years

PRINCIPAL INVESTIGATOR:
Tae-Sic Yoo

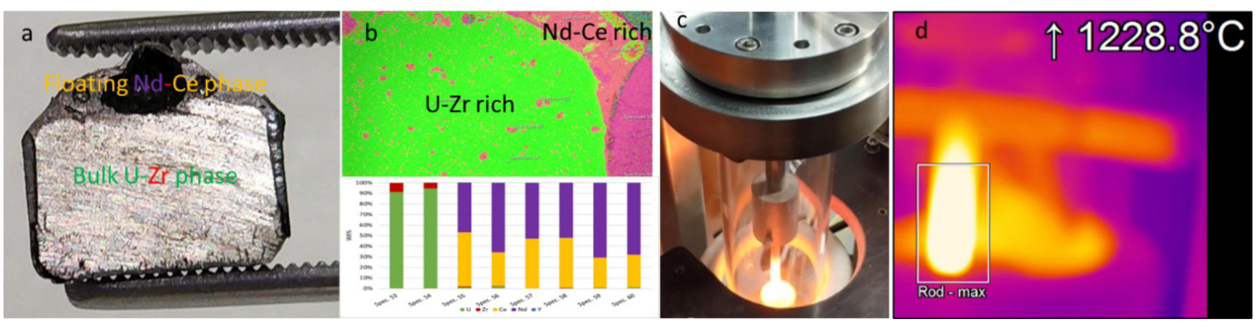
CO-INVESTIGATORS:
Adrian Wagner, INL
Mason Childs, INL

Applying an understanding of fundamental science enables rapid actinide extraction from spent metallic nuclear fuel.

Considering the phase diagrams of metallic spent fuel constituents, the melting and solidifying of spent metallic fuel causes three immiscible layers (actinide-rich, lanthanide-rich, and Group II-rich) and the condensate phase (Group I) to form. This anticipated immiscibility offers an untapped opportunity for innovative fuel cycle solutions. This research confirmed the expected phase behavior and developed a thermal treatment process to rapidly extract actinides from spent metallic fuels. First, melting and solidification experiments with uranium and rare earth metals were performed, and the expected phase separation behaviors between uranium and rare earth metals under a controlled thermal cycle were confirmed. Second, a molten zone system was set up. The system was fully qualified and authorized for experimenting with the quantity of depleted uranium metals for this study and ready for parametric throughput and economic viability study.

The disruptive potential of the project drew attention from funding agencies, and the team secured further research funding from the Advanced Research Projects Agency–Energy: Optimizing Nuclear Waste and Advanced Reactor Disposal Systems program to continue the study regarding transuranics behaviors and economic viability assessment via throughput optimization.

TALENT PIPELINE:
- Mason Childs, postdoc at Texas A&M university, hired as a staff at INL
- Daria Bolgova, student at University of New Mexico



a) Melt consolidated uranium (U)-zirconium (Zr)-cerium (Ce)-neodymium (Nd) sample; b) Scanning electron microscopy analysis of top corner showing clear separation between U-Zr and Ce-Nd phases; c) Inductively heated partially molten U-10Zr rod; d) Real-time temperature measurement with a low wavelength thermal camera.

A Novel Head-end Process for Used Uranium-aluminum Fuels

PROJECT NUMBER:
21A1050-068

TOTAL APPROVED AMOUNT:
$741,000 over 2 years

PRINCIPAL INVESTIGATOR:
Prabhat Tripathy

CO-INVESTIGATORS:
Guy Fredrickson, INL
Michael Patterson, INL
Collin Andersen, University of Utah
Michael Simpson, University of Utah

COLLABORATOR:
University of Texas at San Antonio

Fundamental hydrogen bonding kinetics enable uranium recovery from uranium-aluminum based nuclear fuels.

This project examined the feasibility of developing a dry process to separate aluminum (Al) from used uranium aluminide (UAl_X) fuel. Hydrogen offers the possibility of selectively hydriding uranium present in the uranium aluminide without chemically altering aluminum because aluminum does not form hydrides under the standard conditions of temperature and pressure. Initial experiments involved systematic hydriding test runs involving surrogate aluminides. Two types of surrogate aluminides were subjected to extensive hydriding experiments. These experiments involved heating the alloys to a maximum temperature of 980°C for durations up to 10 hours under positive (100%) hydrogen pressure and continuous pure hydrogen flow conditions. Analysis of the hydrided products indicated the separation of metal hydrides from aluminum metal without the formation of any aluminum hydride phase in the degraded alloys. These promising results formed the basis for repeating the hydriding experiments with the synthetic uranium aluminide alloys. Like surrogate aluminides, uranium aluminides (consisting of a mixture of UAl_2 and UAl_3) showed the early promise for the development of such a dry process. These studies further indicated that although presence of less oxygen is tolerable, significant contamination of the alloys with oxygen can complicate the overall hydriding process. The success of the project, with the help of two university partners, resulted in intellectual property generation and the award of a graduate research fellowship. Additionally, several journal papers are being prepared based on the research.

State of uranium aluminide (UAl_2 + UAl_3) before (left) and after (right) exposure to hydrogen at 700°C for 5 hours.

TALENT PIPELINE:

- Collin Andersen, student at the University of Utah

PRESENTATION:

Anderson, C. T., M. F. Simpson, P. K. Tripathy, "On Hydriding Uranium-Surrogate Aluminides," *American Nuclear Society Winter Meeting and Technology Exposition, Nov. 30–Dec. 3*, Washington DC, USA, 2021.

INTELLECTUAL PROPERTY:

Tripathy, P. K., "Low temperature and inexpensive manufacturing of metal hydride targets," Patent application (Jul. 21, 2021).

AWARD:

This research resulted a 2021 Department of Energy University Nuclear Leadership Program graduate research fellowship for Collin Andersen from the University of Utah.

INTEGRATED ENERGY SYSTEMS

Through research and demonstration, INL delivers technologies advancing integration of energy generation, storage, and delivery needed for a net-zero energy future. INL develops multigeneration energy systems incorporating nuclear heat with all forms of electricity generation, including renewables and natural gas with carbon capture and sequestration, capable of producing products ranging from food to electronic devices as well as power for heating. By innovating methods to harness electricity and heat to produce hydrogen and other industrial products, INL is accelerating the creation of an economy based on clean energy while enhancing grid reliability, resilience, and affordability. INL leverages its core capabilities to demonstrate high-efficiency thermal energy use, enable a sustainable, resilient, and reliable clean energy grid, develop novel chemical and industrial processes, and enhance tools and approaches to optimize integrated energy systems options.


27

Fundamentals for the Valorization of Carbon Dioxide Toward Integration to Nuclear Power Plants

PROJECT NUMBER:
20A44-083

TOTAL APPROVED AMOUNT:
$1,801,000 over 3 years

PRINCIPAL INVESTIGATOR:
M. M. Ramirez-Corredores

CO-INVESTIGATORS:
Christopher Zarzana, INL
Luis Diaz Aldana, INL

Gamma radiation induced conversion of carbon dioxide and methane to carbon monoxide and hydrogen is a cost-effective, negative emissions process technology.

Carbon dioxide (CO_2) emissions are a significant contributor to anthropogenic climate change. Conversion of CO_2 into value-added products can contribute to economic sustainability when implemented through a circular economy. Because CO_2 conversion reactions require significant amounts of energy, low carbon emitting sources like nuclear energy must be used as part of a sustainable decarbonized future. Ionizing radiation as well as excess heat, electricity, or both generated by a nuclear power plant were considered for driving CO_2 conversion into syngas, a mixture of hydrogen and carbon monoxide. This project examined the fundamental radiation chemistry of mixtures of CO_2 and methane for visualizing the potential interactions of intermediate products with catalysts or electrocatalysts and their role on guiding propagation or termination reactions into desirable products. The outputs from the project include

1. An irradiation gas reactor cell,
2. Electrochemical baseline tests determining,
   a. The effect of gamma radiation on the components of the electrochemical cell,
   b. The level of CO_2 conversion in the absence of radiation, and
   c. Electrochemical conversion of CO_2 to carbon monoxide in the absence of radiation,
3. Catalyst functionality results and potential formulations preparations,
4. Catalytic baseline tests,
5. Systematic study of the effect of dose in the radiolysis of CO_2, methane and their 1:1 mixture, and
6. Reaction under irradiation in the presence of catalysts tests.

The research results can lead to technology development for waste carbon utilization and more efficient energy usage from nuclear power plants, which will close the carbon cycle, create new jobs, and open a $50 billion market.

TALENT PIPELINE:

- Elizabeth Parker-Quaife, postdoc at INL
- Ryan Morco, postdoc at INL

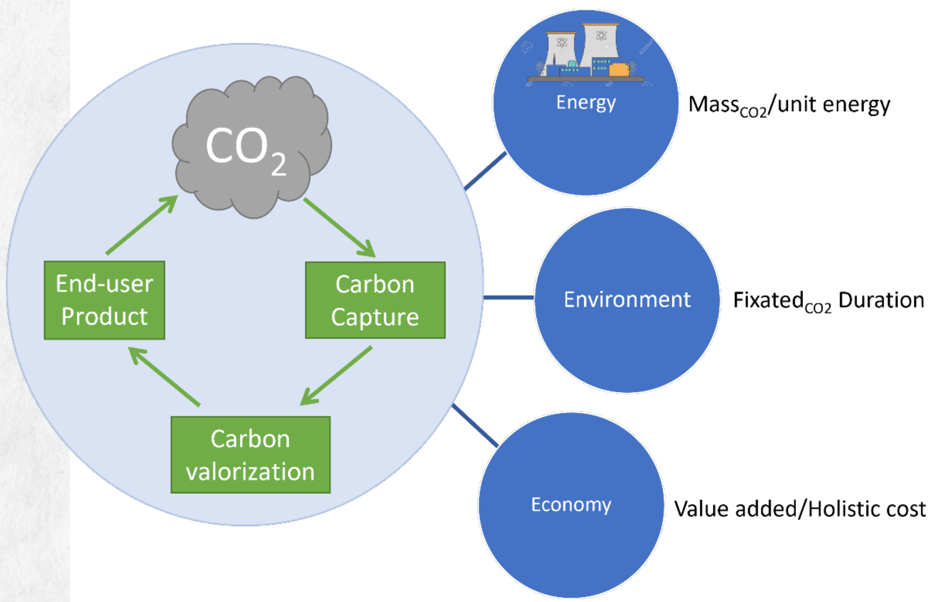
PUBLICATIONS AND PRESENTATION:

Ramirez-Corredores, M. M., G. Gadikota, E. E. Huang, A. M. Gaffney, "Radiation induced chemistry of carbon dioxide: A pathway to close the carbon loop for a circular economy," *Frontiers in Energy Research* 8 (108) (2020) 17.

Ramirez-Corredores, M. M. L. A. Diaz, A. M. Gaffney, C. A. Zarzana, "Identification of opportunities for integrating chemical processes for carbon (dioxide) utilization to nuclear power plants," *Renewable and Sustainable Energy Reviews* 150(111450) (2021) 15.

Ramirez-Corredores, M. M., "Carbon dioxide valorization: A needed step within a decarbonization strategy," *Proc. Global Experts Meet on Chemical Engineering and Technology*, Las Vegas, NV. USA, Jul. 14–15 (2022) 15.

Ramirez-Corredores, M. M., M. R. Goldwasser, E. F. D. S. Aguiar, "Decarbonization as a route toward sustainable circularity." *1 ed. Springerbriefs in Applied Sciences and Technology*, ed. Doyle, A. 2023, Switzerland AG: Springer Cham. 164 (+XVI) Dec. 16, 2022.



Valorization of CO_2 as a step toward sustainably closing the carbon cycle

Improving the Stability and Durability of Aptamer Based Smart Materials Using Extremophiles

PROJECT NUMBER:
20A44-203

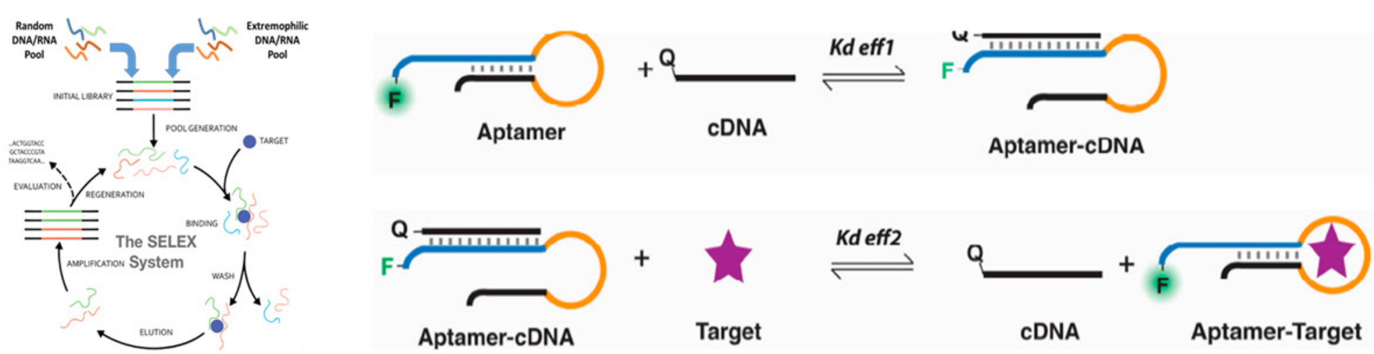
TOTAL APPROVED AMOUNT:
$550,000 over 3 years

PRINCIPAL INVESTIGATOR:
Lorenzo Vega-Montoto

CO-INVESTIGATORS:
Dayna Daubaras, INL
Caryn Evilia, Idaho State University

Smart biomolecules can be used to create materials and sensors for environmental, advanced manufacturing, and homeland security applications.

There is an increasing necessity to develop a standardized, cost-effective, robust, and adaptable methodology to produce smart molecules that can detect, interact, and respond to a diverse variety of chemical and biochemical compounds selectively and sensitively. In addition, these smart molecules should be characterized by their flexibility, robustness, resilience, and durability because they will be most likely used in very extreme environments for long time periods. Nucleic acid and peptide aptamers are smart molecules that can dose respond with high levels of sensitivity and specificity to target molecules ranging from simple ions to complex whole cells. Even though aptamers are more chemically stable than antibodies, their stability and durability might not be enough for extreme environments or long periods. Post-production chemical modifications have improved stability, but they might compromise sensitivity and specificity. This project established the basis for a simple, robust solution to improve the stability and durability issues of these flexible smart molecules by starting the selection process with a library of proteo-geno-transcriptomic material from extremophiles that have been proven to withstand extreme conditions instead of a library of random sequences. The Systematic Evolution of Ligands by Exponential procedures were developed and optimized to produce aptamers from random and extremophile derived libraries. Quality control methodologies to assess the progress and completion of the Systematic Evolution of Ligands by Exponential methodology as well as bioinformatic pipelines to perform next generation sequencing analysis and motif analysis were implemented to decide the most promising aptamers for each target. These studies led to the identification of promising aptamer candidates to perform exploratory affinity and specificity studies to assess the intrinsic selectivity and specificity that will be required to develop smart material applications.



Systematic Evolution of Ligands by Exponential approach allows the identification of aptamers that can be used to develop dose dependent, selective and specific fluorescent sensors.

TALENT PIPELINE:

- Skyler Smith, student at Idaho State University
- Alex Guerrero, student at Idaho State University
- Jack Carmack, student at University of Oklahoma

PRESENTATIONS:

Smith S., A. Guerrero, D. Daubaras, L. E. C. Vega-Montoto, "Utilizing extremophile DNA within SELEX to select for aptamers that bind metal ions," *ACS Spring Meeting*, Apr. 5–May 1, 2021.

Vega-Montoto, L., D. Daubaras, S. Smith, K. Schaller, S. Smith, A. Guerrero, D. Daubaras, C. Evilia, "Improving the Stability and Durability of Aptamer Based Smart Materials Using Extremophiles," *Federation of Analytical Chemistry and Spectroscopy Societies*, Sep. 26–Oct. 1, 2021.

Data-driven Failure Diagnosis and Prognosis of Solid-state Ceramic Membrane Reactor under Harsh Conditions Using Deep Learning Technology with Internal Voltage Sensors

PROJECT NUMBER:
21A1050-090

TOTAL APPROVED AMOUNT:
$800,000 over 2 years

PRINCIPAL INVESTIGATOR:
Wei Wu

CO-INVESTIGATORS:
Congjian Wang, INL
Dong Ding, INL

COLLABORATOR:
Center for Advanced Energy Studies

Machine learning based on real-time electrochemical data predicts solid-state ceramic membrane reactor service time.

The solid-state ceramic membrane reactor concept can play an important role in the energy-to-molecules/materials pillar by using energy associated with renewables and nuclear to manufacture functional intermediates, fuels, and chemicals. One of technical challenges is the difficulty in real time characterizing and predicting solid-state ceramic membrane reactor failure mechanisms because they operate in harsh environments, including, elevated temperatures, various gas compositions (high steam, hydrocarbons, hydrogen, etc.), corresponding reactions, and so on. More importantly, quantitative understanding of different solid-state ceramic membrane reactor component contributions to the total degradation under realistic operating conditions is extremely hard to obtain, but critical to illuminate the long-term performance and develop mitigations to materials failure.

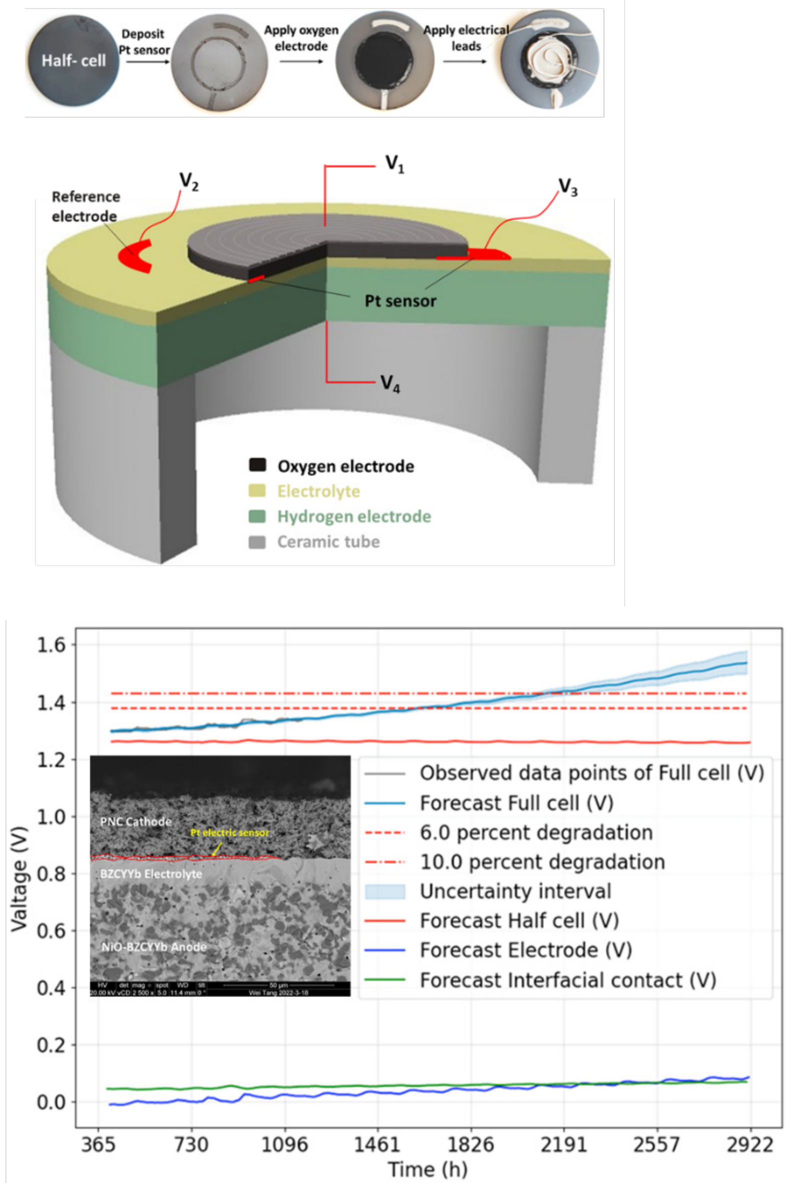
In this project, a reliable data-driven modeling method was developed for in situ investigation of different component effects (anode, electrolyte, and cathode) on the degradation behavior in a solid-state ceramic membrane reactor. This was accomplished by embedding micro-voltage sensors into the interface between different components to monitor current response and to collect impedance electrochemical data during operation. This method was applied to predict the protonic ceramic electrochemical cells lifetime when used in high temperature steam electrolysis to produce hydrogen. By analyzing the electrochemical results via machine learning, a Bayesian calibration tool was developed and implemented to calibrate the simulation models for the failure analysis and lifetime prediction. This tool uses real-time electrochemical data to exhibit performance degradation. The data are generated from in-house fabricated platinum electric interfacial sensors embedded in a protonic ceramic electrochemical cell under steam electrolysis conditions. This approach highlights the promise of combining data generation and data-driven machine learning to understand and develop complex systems. The approach also reveals the underlying degradation mechanism, the understanding of which can assist in developing relevant mitigation strategies. Such mitigation strategies are essential to the technology market penetration and commercialization.

TALENT PIPELINE:

- Wenjuan Bian, postdoc at INL
- Bin Hua, postdoc at INL

PRESENTATION AND PUBLICATION:

Wu, W., W. Bian, J. Li, D. Ding, "Revitalizing performance and expanding lifetime of protonic ceramic cells by interfacial acid etch," Oral presentation, *23rd International Conference on Solid-state Ionics*, Jul. 17–22, Boston, MA, USA, 2022.



Interfacial electrical sensor enables root cause analysis for ceramic membrane reactor degradation, thus increase the validity of simulation.

Natural Gas to Chemical Intermediate by Single Step Synthesis Using Tubular Ceramic Catalytic Membrane Reactor

PROJECT NUMBER:
21A1050-119

TOTAL APPROVED AMOUNT:
$800,000 over 2 years

PRINCIPAL INVESTIGATOR:
Hanping Ding

CO-INVESTIGATORS:
Dong Ding, INL
Hongqiang Hu, INL
Haiyan Zhao, University of Idaho

COLLABORATORS:
George Mason University
University of Maine
Kansas State University

Directly converting natural gas to aromatics and hydrogen can reduce the environmental impact of the petrochemical industry.

Aromatics like benzene, toluene, and xylenes are the essential building blocks for some of the most used petrochemical products. The vast majority (97%) of today's aromatics production relies on crude oil. The supply of aromatics mainly hinges on three different sources: steam cracking of naphtha, catalytic reforming, and coke-oven light oil. All three processes require the production of significant amounts of synthesis gas, in turn creating significant amounts of carbon dioxide and leading to high capital and production costs. Methane, the main component of natural gas, has the highest hydrogen to carbon ratio of all hydrocarbons. Therefore, it is more environmentally friendly in terms of carbon dioxide emissions than oil or coal-derived fuels. Furthermore, North America and methane hydrate in the sediments of the ocean floors are conservatively estimated to represent twice the amount of carbon in all other known fossil fuel reserves. An efficient methane dehydroaromatization catalyst should i) effectively activate stable carbon-hydrogen bonds in methane molecules, ii) allow selective production of light aromatics, minimizing the formation of unwanted graphitic carbon (coke), and iii) remain stable at high reaction temperatures.

Two directions were chosen to achieve the objectives. First, a new ternary catalyst was designed in a fixed-bed reactor—molybdenum-based catalyst using bimetallic platinum-bismuth as a promoter—that shows a 28% increased methane conversion and higher selectivity to benzene compared to conventional molybdenum-based catalyst. In addition, the catalyst was integrated with a proton conducting membrane reactor to cogenerate aromatics and electricity, which showed remarkable performance. It presented a 20% single-pass methane conversion and 35% benzene yield in the membrane reactor, as well as a high output power density of 275 $mW \cdot cm^{-2}$ in methane at 700°C.

TALENT PIPELINE:

- Pengxi Zhu, student at George Mason University
- Mark Hull, student at University of Idaho

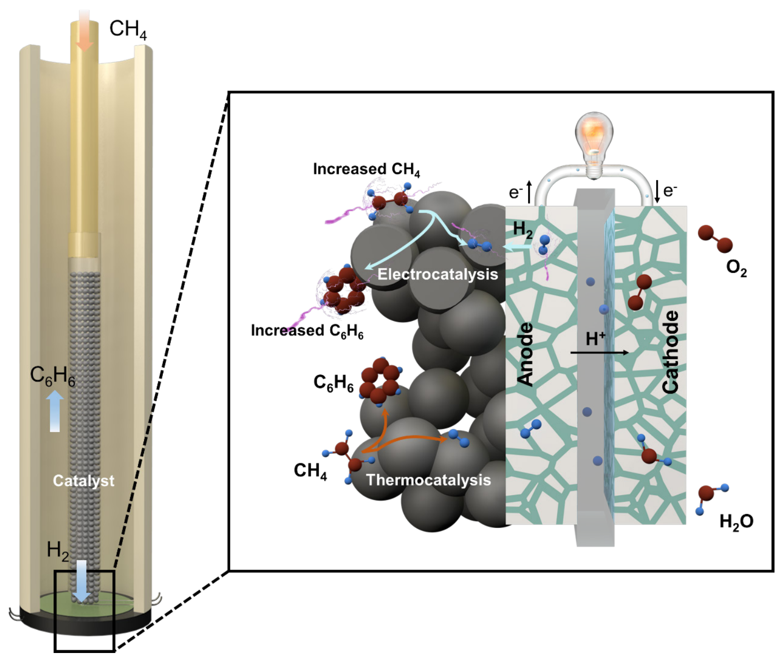
PUBLICATION AND PRESENTATIONS:

Zhu, P., L.-C. Wang, F. Stewart, D. Ding, J. Matz, P. Dong, H. Ding, "Direct Conversion of Natural Gases in Solid Oxide Cells: A Mini-Review," *Electrochemistry Communications* (2021) 128.

Ding, H., D. Ding, "Advanced Electrode and Electrolyte Materials for Proton Conducting Solid Oxide Electrolysis Cells," *241st Electrochemical Society Meeting* (2022) Spring, Vancouver Canada (Invited talk).

Ding, H., P. Zhu, P. Dong, D. Ding, "Direct Conversion of Natural Gases to Aromatics in Solid Oxide Membrane Reactor," oral presentation, *240th Electrochemical Society Meeting Virtual*, 2021 Fall.

Ding, H., P. Zhu, P. Dong, D. Ding, "Natural Gas Conversion Using Proton Conducting Ceramic Membrane Reactor," oral presentation, *239th Electrochemical Society Meeting Virtual*, 2021 Spring.



Schematic of the working principle where methane (CH_4) is converted to benzene (C_6H_6) and hydrogen (H_2) via a platinum-copper/molybdenum/zeolite catalyst. H_2 is transported as protons (H^+) to the cathode side and generates electricity.

Up-cycling Process Feasibility for Coupled Radiolytic and Biochemical Conversion of Polyethylene

PROJECT NUMBER:
21A1055-015

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PRINCIPAL INVESTIGATOR:
Gregory Horne

CO-INVESTIGATORS:
Corey Pilgrim, INL
Julie Peller, Valparaiso University
Michael Watters, Valparaiso University
Stephen Mezyk, California State University Long Beach

Biochemical digestion of gamma irradiated polyethylene by the fungus Neurospora crassa in anaerobic saltwater conditions is not practical.

There are currently no sustainable industrial scale processing routes for the recovery and conversion of low value plastics such as polyethylene. Therefore, they accumulate in landfill waste and contaminate the environment. Due to the absence of sustainable recycling methods, the demand for virgin plastic synthetic feedstocks from petrochemical sources continues to increase and is expected to become a significant energy drain by 2050. From both an energy and environmental standpoint, this unsustainability has prompted DOE to encourage the development of sustainable plastic recycling methods because the billions of tons of waste plastic are potential sources of cost-effective synthetic feedstocks. However, unlike several other types of plastic, such as polyesters, polyethylene does not contain any chemical functionality beyond its carbon-hydrogen backbone, essentially making it chemically and biologically inert unless extreme conditions or exotic processes are employed. The research presented here investigated the proof-of-concept of using gamma irradiation capabilities at INL to initiate the chemical functionalization and depolymerization of polyethylene micro- and nanoplastics blended into aqueous solutions (water and saltwater), followed by biochemical conversion at Valparaiso University to yield useful synthetic feedstocks. This research found that anaerobic conditions and saltwater were not practical for a radiation-driven polyethylene up-cycling process. However, the anerobic data gathered by this work does provide a baseline for future investigations into the practicality and impact of aerobic conditions. Biochemical conversion studies using the fungus Neurospora crassa found that non-irradiated and anaerobic irradiated polyethylene was unavailable as a carbon source and mostly toxic to the microbes. However, the presence of more functionalized polymers, such as polyethylene terephthalate, afforded viable Neurospora crassa cultures. These latter findings present a promising pathway should aerobic irradiations lead to carbonyl functionality in polyethylene. This work also developed an effective treatment for commercially procured polyethylene to remove residual contaminants, the publication of two peer reviewed manuscripts, dissemination of results through eight presentations, mentoring of 11 students, and the successful award of follow-on funding through the National Science Foundation.

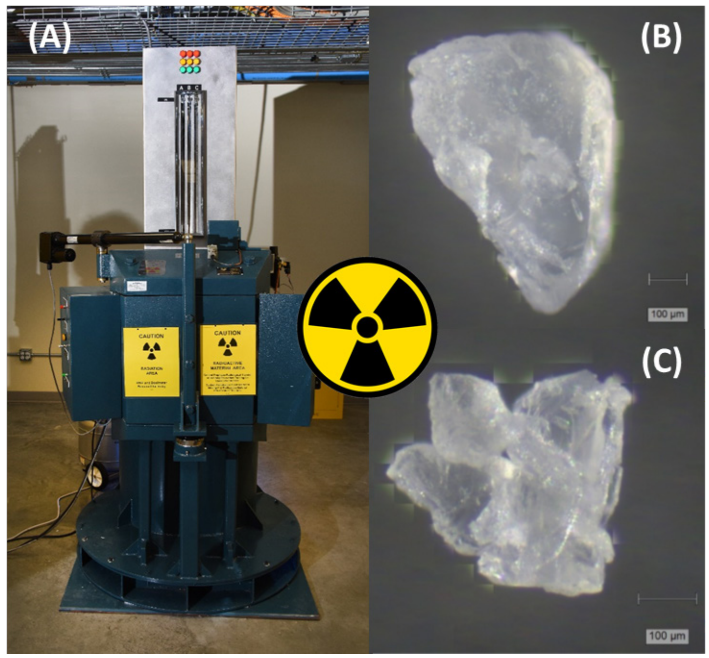
TALENT PIPELINE:

- Joe Castleman, student at Valparaiso University
- Kayla Cheney, student at California State University Long Beach
- Richard F. Faulkner, student at California State University Long Beach
- Scott Kaiser, student at Valparaiso University
- Morgan Keller, student at Valparaiso University
- Seth Junglas, student at Valparaiso University
- Sydney Martens, student at Valparaiso University
- Megan Moretti, student at Valparaiso University
- Brian Tran, student at California State University Long Beach
- Heather Wendland, student at Valparaiso University
- Antigone Wilson, student at Valparaiso University

PUBLICATIONS:

Peller, J. R., S. P. Mezyk, S. Shidler, J. Castleman, S. Kaiser, G. P. Horne, "The reactivity of polyethylene microplastics in water under low oxygen conditions using radiation chemistry," *MDPI Water* 13 (2021) 3120.

Peller, J. R., S. P. Mezyk, S. Shidler, J. Castleman, S. Kaiser, R. F. Faulkner, C. D. Pilgrim, A. Wilson, S. Martens, G. P. Horne, "Facile nanoplastic formation from macro and microplastics in aqueous media," *Environmental Pollution* 313 (2022) 120171.



(A) INL Center for Radiation Chemistry Research high dose rate Foss Therapy Services cobalt-60 gamma irradiator, within which polyethylene microplastics were irradiated in anaerobic aqueous solutions and subsequently analyzed by dark field microscopy. (B) non-irradiated polyethylene particle, and (C) post-high-dose gamma irradiated polyethylene particle.

Flexible Chemical Manufacturing for Carbon Dioxide Hydrogenation to Methanol

PROJECT NUMBER:
21P1062-015

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PRINCIPAL INVESTIGATOR:
Daniel Ginosar

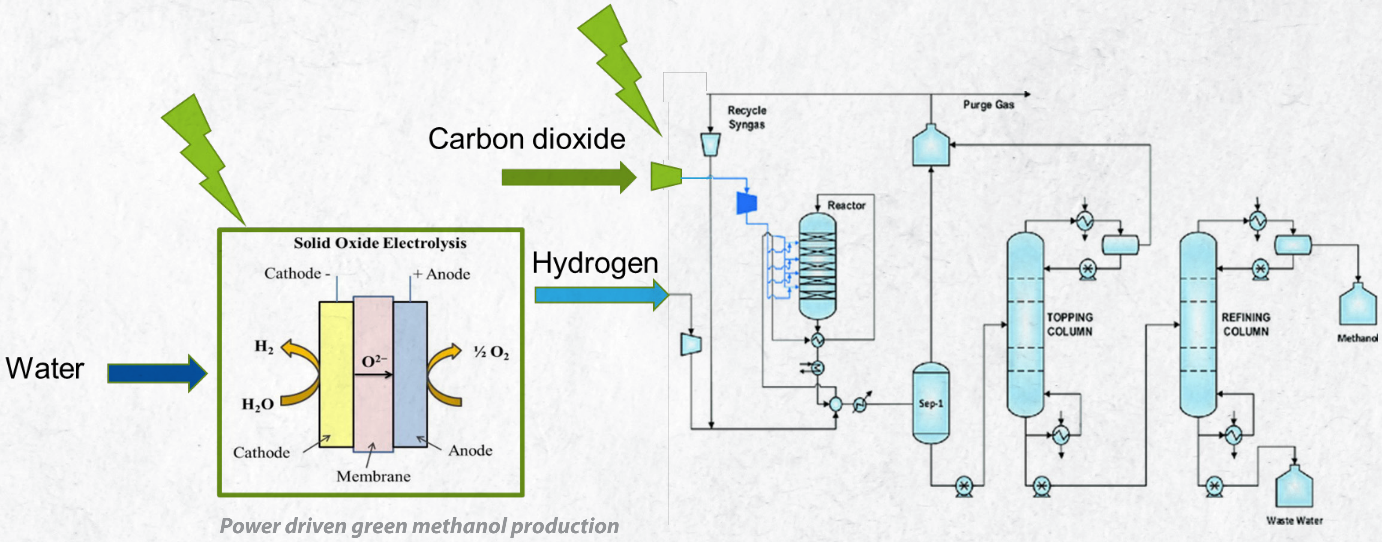
CO-INVESTIGATORS:
Binghui Li, INL
Birendra Adhikari, INL
Rebecca Fushimi, INL
Yixiao Wang, INL

Flexible chemical manufacturing plant operation can result in economical production of environmentally sustainable methanol.

Issues surrounding grid stability have become more significant as more and more variable renewable energy resources have come online. The greater contributions of variable renewable energy to the power grid have led to power price volatility, causing market marginal hourly prices to peak at hundreds of dollars per megawatt hour and drop to net negative values. Operating flexible chemical manufacturing plants at full speed when prices are low or negative and minimizing the production processes when power prices are high is a way to capitalize on these power price volatilities. More importantly, these operations can have net-zero carbon emissions by capitalizing on the availability of low carbon power sources. The feasibility of flexible chemical manufacturing was examined to produce green methanol: the reaction of CO_2 with hydrogen where the hydrogen is obtained from water electrolysis. The operation is powered by low carbon energy sources such as nuclear, solar, and wind. Flexible chemical manufacturing can be employed to essentially convert water and CO_2 into a valuable fuel or chemicals when power production exceeds demand and minimized when demand exceeds supply. To test this concept, the research team constructed and tested a variable flow pressurized CO_2 hydrogenation reactor system using an economic and environmental driven flow algorithm to understand the performance and stability of a commercially available copper/zinc oxide/alumina catalyst. Results showed that for one hundred hours of continuous operation at a space velocity of 6,000 h^{-1} at a pressure of 50 bar and a reactor inlet temperature of 230°C, the catalyst activity was stable.

TALENT PIPELINE:

- Amit Nilkar, student at University of Idaho



Power driven green methanol production process flow diagram.

Advanced Materials and Manufacturing for Extreme Environments

Building on existing expertise in materials research and development for extreme environments with innovation in advanced manufacturing, INL is shifting the paradigm from design-build-test to digital design and manufacturing for nuclear fuels, lightweight materials, and advanced survivability materials. Advanced manufacturing enables simultaneous fabrication with process monitoring and control, and supports all INL mission area needs. Under this initiative, INL uses its core capabilities to accelerate innovation by developing advanced manufacturing process-informed material design, expand advanced manufacturing process development, enable rapid material characterization and testing designed for advanced manufacturing, and integrate comprehensive data analytic and modeling and simulation techniques.


CAUTION

Multi-role High Entropy Alloys for Extreme Environments

PROJECT NUMBER:
20A44-011

TOTAL APPROVED AMOUNT:
$885,500 over 3 years

PRINCIPAL INVESTIGATOR:
Subhashish Meher

CO-INVESTIGATORS:
Guoping Cao, INL
Michael Glazoff, INL
Prabhat Tripathy, INL
Thomas Lillo, INL

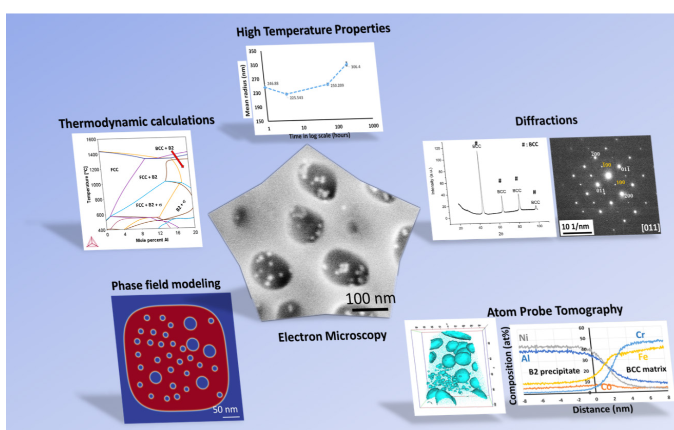
Novel microstructural design and manufacturing of high entropy alloys establish a new paradigm for developing materials that operate in thermal and mechanical extreme environments.

Nuclear, fossil, and solar power generation systems need materials capable of operating for several decades while maintaining adequate strength at temperatures approaching or exceeding 800°C. The emerging class of high entropy alloys have the potential to be tailored for these extreme environments. This project implemented a calculation of phase diagram based approach to accelerate exploration of multi-role high entropy alloy compositions that would exhibit desired microstructure properties for basic fuel saving (fossil), output application (nuclear co-generation), and making systems technically feasible (solar). Novel solid-state transformation pathways such as structural hierarchy in ordered precipitates have been explored in a body-centered cubic based high entropy alloy via coupled advanced characterization and phase field modeling. The results suggest that the high temperature precipitate coarsening can be delayed significantly due to structural hierarchy. Also, the intriguing aspect of selective coarsening and dissolution of ordered precipitates in high entropy alloys when subjected to high temperatures was understood via advanced microscopy and phase field modeling. This project utilized state-of-the-art characterization and experimental facilities at INL and Center for Advanced Energy Studies to understand the collective mechanical response of high entropy alloys under coupled thermal and mechanical extremes. Furthermore, high entropy alloy manufacturing from oxide powder mixtures was demonstrated via a novel cost-effective electro-deoxidation method. This new experimental capability will be essential for the future scaling of this process for larger production of alloy powders. The exploration of high temperature high entropy alloys with novel microstructural design and manufacturing provides a solid foundation for a new paradigm of alloy design required for operation in coupled thermal and mechanical extremes associated with advanced nuclear reactors.

TALENT PIPELINE:

- Sanu Gupta, student at Oregon State University
- Sourabh Kadambi, postdoc at INL

A representation of coupled experimental and modeling implemented in the project for physical understanding of high entropy alloys.



Direct Integrated Advanced Manufacturing of Multi-component Solid Fuel Systems with Embedded Sensors

PROJECT NUMBER:
20A44-131

TOTAL APPROVED AMOUNT:
$774,500 over 3 years

PRINCIPAL INVESTIGATOR:
Elizabeth Zell

CO-INVESTIGATORS:
Isabella Van Rooyen, INL
Sudipta Biswas, INL
Edward Herderick, The Ohio State University

A binder-free slurry of ceramic nuclear fuel surrogate formed free-standing shapes, advancing additive manufacturing of reactor materials.

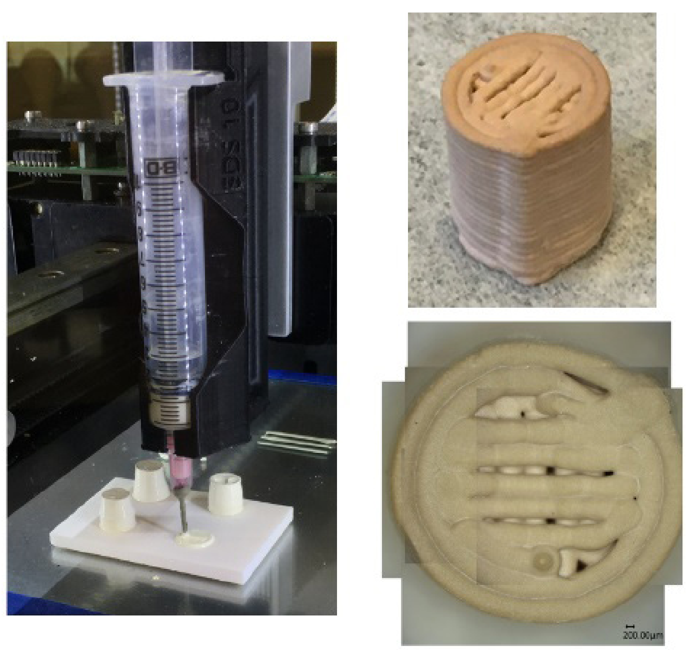
This project developed a sol-gel-like additive manufacturing process for high temperature fuel and cladding applications through collaboration between INL and The Ohio State University Center for Design and Manufacturing Excellence. Extrusion of slurry via syringe followed by drying and sintering allowed for direct printing of ceramic materials. Cerium dioxide, a surrogate for nuclear fuel and a focus of this work, was incorporated in a binder-free slurry, leaving printed parts without polymer or other residual additives. The developed slurry was extruded to build free-standing cylinders of 20 layers and 9 mm height. The cylinders were subsequently dried in ambient air and sintered at 1500°C for one hour. Titanium dioxide slurries were also printed because the material is applicable in gas sensing. Furthermore, cerium dioxide was printed within a silicon carbide bath, thus incorporating the fuel surrogate in a cladding material and approaching multi-material printing. In future work, printing in a bath could support uncured or low modulus gels that cannot stand freely or otherwise require a support bath, thus improving the applicability of additive manufacturing to ceramic fuels, claddings, and other materials and multi-material systems. There is also future potential to include embedded sensor technologies and new fuel-gap fabrication technologies.

TALENT PIPELINE:

- Elizabeth Zell, postdoc at INL
- Jakub Toman, postdoc at INL

PRESENTATIONS

Zell, E., E. D. Herderick, I. V. Rooyen, "A Review of Solution Based Processing Routes for Advanced Nuclear Fuel," *The Minerals, Metals and Materials Society 2021* Mar. 15–18, 2021.



Layer-by-layer extrusion three-dimensional printing of nuclear fuel surrogate ceramic and views of final, sintered product.

Advanced Manufacturing of Heat Pipes with Connectivity to Thermoelectrics

PROJECT NUMBER:
20A44-065

TOTAL APPROVED AMOUNT:
$1,084,000 over 3 years

PRINCIPAL INVESTIGATOR:
Donna Guillen

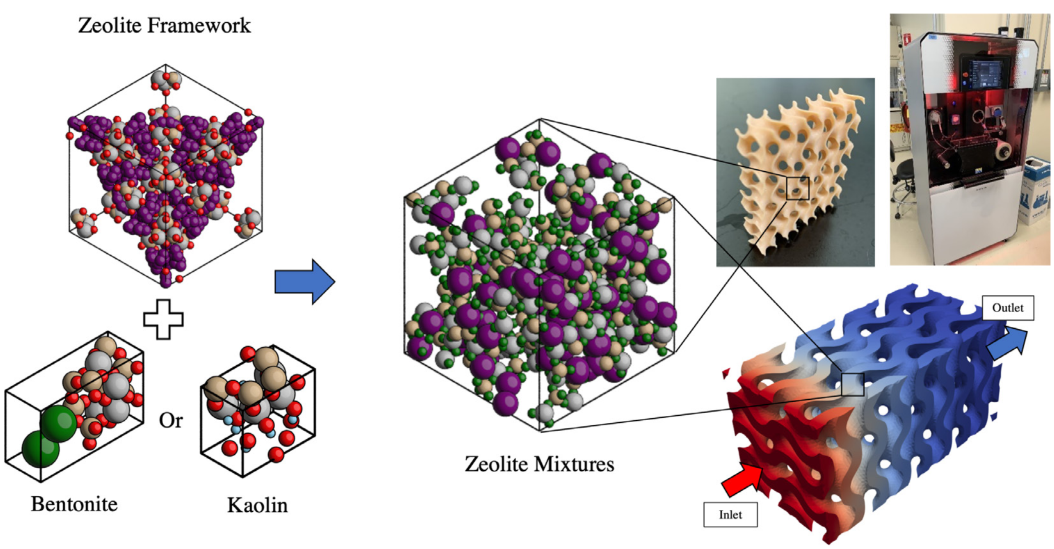
CO-INVESTIGATORS:
Adrian Wagner, INL
Piyush Sabharwall, INL
Troy Unruh, INL

COLLABORATORS:
Boise State University
Northwestern University

Researchers combined advanced computational modeling, digital design, and three-dimensional printing to create high-performance heat pipes, heat exchangers, and thermoelectric devices.

Thermoelectric devices are solid-state devices used to convert thermal energy to electrical energy. However, current deployment of these devices is limited by high manufacturing costs and low efficiency. Integrating a heat pipe heat exchanger with the thermoelectric platform can eliminate thermal contact resistances and increase device performance. Additive manufacturing of the heat exchanger enables the fabrication of a compact device with an optimized configuration that cannot be readily produced by traditional manufacturing methods. The heat exchanger topology features a triply periodic minimal surface that offers the potential for lighter weight structures. The hot side of the heat exchanger consists of a triply periodic minimal surface printed in 316L stainless steel using a laser powder bed fusion process, whereas the cold side of the heat exchanger features a triply periodic minimal surface structure printed with 13X zeolite using a digital light processing printer. Both laser powder bed fusion and digital light processing printers construct the component in a layer-by-layer manner. Heat pipes transfer heat through the triply periodic minimal surface structure to the thermoelectric devices.

The n-type titanium-nickel-tin thermoelectric legs were manufactured via an additive manufacturing process of ink-extrusion, whereas the p-type legs were fabricated by spark plasma sintering. The additive manufacturing process for creating the half-Heusler titanium-nickel-tin thermoelectrics consisted of ink printing titanium nickel lattices, debinding and thermal sintering, and infiltrating tin. The technique was developed in collaboration with Northwestern University.



Zeolite binder integration using either bentonite or kaolin. Inset upper right: digital light processing printer used to fabricate an optimized geometry for adsorption.

TALENT PIPELINE:

- Miu Lun Lau, student at Boise State University
- Patrick Moo, student at Florida State University
- Nathan Linton, student at University of Wyoming
- Laura Ziegler, student at University of Utah
- Kari Perry, student at Montana State University
- Alexander Proschel, student at Northwestern University

A thermoelectric device was constructed and tested to demonstrate the performance of the heat pipe heat exchanger thermoelectric platform. The heat pipes, heat exchanger, and thermoelectric components were modeled in MOOSE to optimize their thermal and electrical performance. The novel integration of the heat pipe, heat exchanger, and thermoelectric device can be applied to a myriad of thermal transport applications, including wind, solar and nuclear energy, waste heat recovery, and other heat removal systems.

PUBLICATION AND PRESENTATIONS:

Guillen, D. P., "Machine Learning Applications in Advanced Manufacturing Processes," *Journal of the Minerals, Metals and Material Society* 72(11) (2020) 3906–3907.

Guillen, D. P., A. Wagner, P. Moo, C. Turner, "Additive Manufacturing of Heat Pipes," *2020 The Minerals, Metals, and Materials Society Meeting*, Feb. 23–27, 2020.

Guillen, D. P., P. Moo, M. Shaltry, R. O'Brien, "Advances in Digital Light Processing Printing for Energy Applications," *2021 The Minerals, Metals and Materials Society Meeting*, Mar. 14–18, 2021.

Proschel, D. P. Guillen, D. Tucker, D. Dunand, "3D Ink-Extrusion Printing of Thermoelectric Materials onto Heat Exchangers," *2022 The Minerals, Metals and Materials Society Meeting*, Feb. 27–Mar. 3, 2022.

Additive Manufacturing of Multi-functional, Large-scale Components

PROJECT NUMBER:
20A44-113

TOTAL APPROVED AMOUNT:
$796,500 over 3 years

PRINCIPAL INVESTIGATOR:
Thomas Lillo

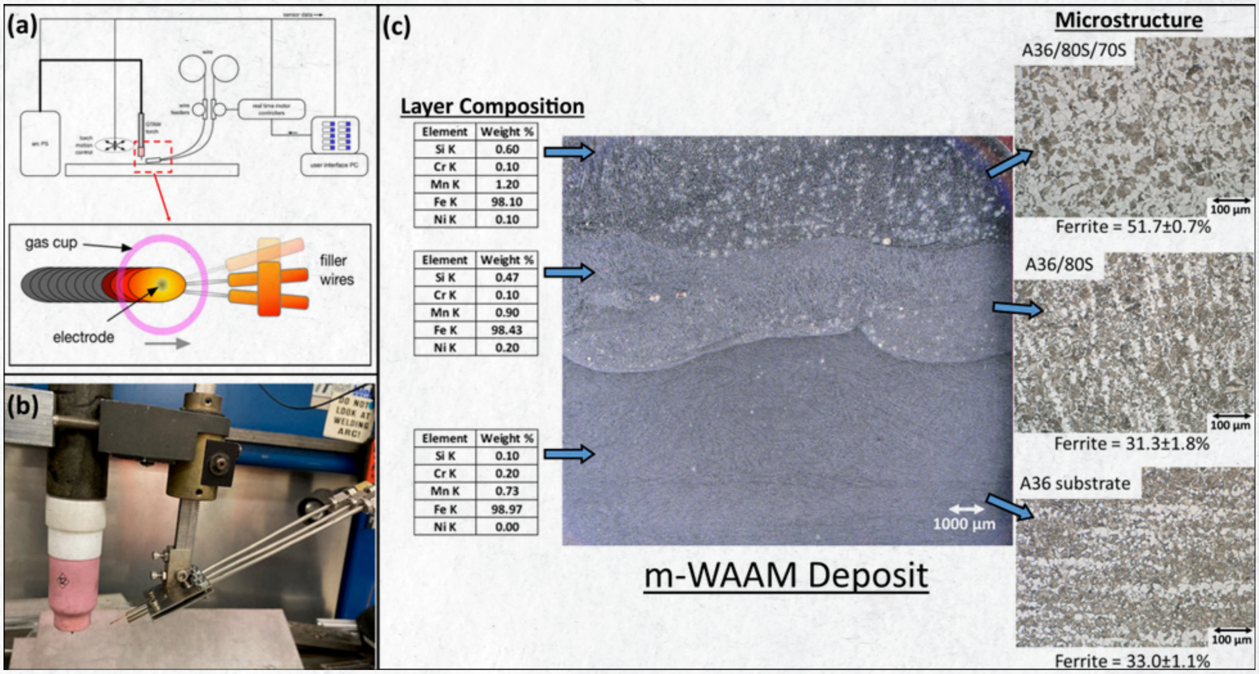
CO-INVESTIGATORS:
Eric Larsen, INL
Jason Walleser, INL
Michael Glazoff, INL
Nathan Huft, INL
Denis Clark, DEClark Welding Engineering, PLLC

A multi-wire, arc-based, metal additive manufacturing system rapidly builds functionally graded components for harsh environments.

Functionally graded components offer the potential to address materials requirements in harsh environments on a local scale and eliminate the need to develop a metallic alloy that satisfies all design requirements, such as physical property requirements like high temperature structural strength, thermal conductivity, etc., as well as environmental resistance criteria. Researchers developed an additive manufacturing system that can deposit controlled compositions on a local scale during component fabrication. The multi-wire arc additive manufacturing system can deliver up to three different welding wires into the weld pool to control the local alloy composition, effectively placing optimum alloy compositions where they are needed in a component. For example, a component could be manufacturing with corrosion resistant alloy internally with high strength structural alloys on the outside. Because the system is based on arc welding, high deposition rates (approximately 10-100 times that of powder-based three-dimensional metal printing) and high material utilization efficiency (near 100% for multi-wire arc additive manufacturing versus approximately 40-60% for powder-based three-dimensional metal printing) can be achieved.

Researchers demonstrated that deposits could be continuously graded from one end of the build to the other. Simple walls were fabricated that were graded from a high temperature nickel-based structural alloy, Hastelloy N, to pure nickel, which is highly resistant to molten salts but lacks any significant high temperature structural strength. Furthermore, it was demonstrated that the characteristics of the composition gradient in layer-by-layer builds could be influenced by deposition parameters, such as heat input, ranging from a stepped profile at low heat input to an almost continuous gradient at higher heat inputs due to the extent of dilution from the underlying layer as influenced by heat input. Finally, it was demonstrated that design of the proper composition gradient in conjunction with a proper post-build heat treatment could produce a gradient in microstructure and properties in simple iron-based builds. The influence of these graded microstructures on shock wave propagation was quantified using dynamic compression tests using the split Hopkins pressure bar system.

This project just scratches the surface of what is possible with multi-wire arc additive manufacturing and starts to elucidate the influence of the numerous operating parameters. Also, some of the design considerations and issues associated with building large functionally graded components are now evident. However, the advantages of multi-wire arc additive manufacturing in joining dissimilar metals and applying corrosion resistant claddings to high temperature structural metals are immediately clear from the research results.



(a) Schematic of the multi-wire arc additive manufacturing (m-WAAM) system, (b) multi-wire guide tubes and (c) m-WAAM deposit with a gradient in composition and microstructure.

TALENT PIPELINE:

Tate Patterson, student at The Ohio State University, hired at INL

PRESENTATION:

Lillo, T. M., N. J. Huft, M. V. Glazoff, J. A. Simpson, D. E. Clark, "Novel Aspects of multi-Wire Arc Additive Manufacturing for Large Component Fabrication for Extreme Environments and New Alloy Discovery," *The Minerals, Metals and Materials Society 2021 Annual Meeting and Exhibition*, Mar. 15–18, 2021.

Development of a Laser Ultrasonics-based Approach for Rapid Screening of High Entropy Alloys

PROJECT NUMBER:
21A1055-026

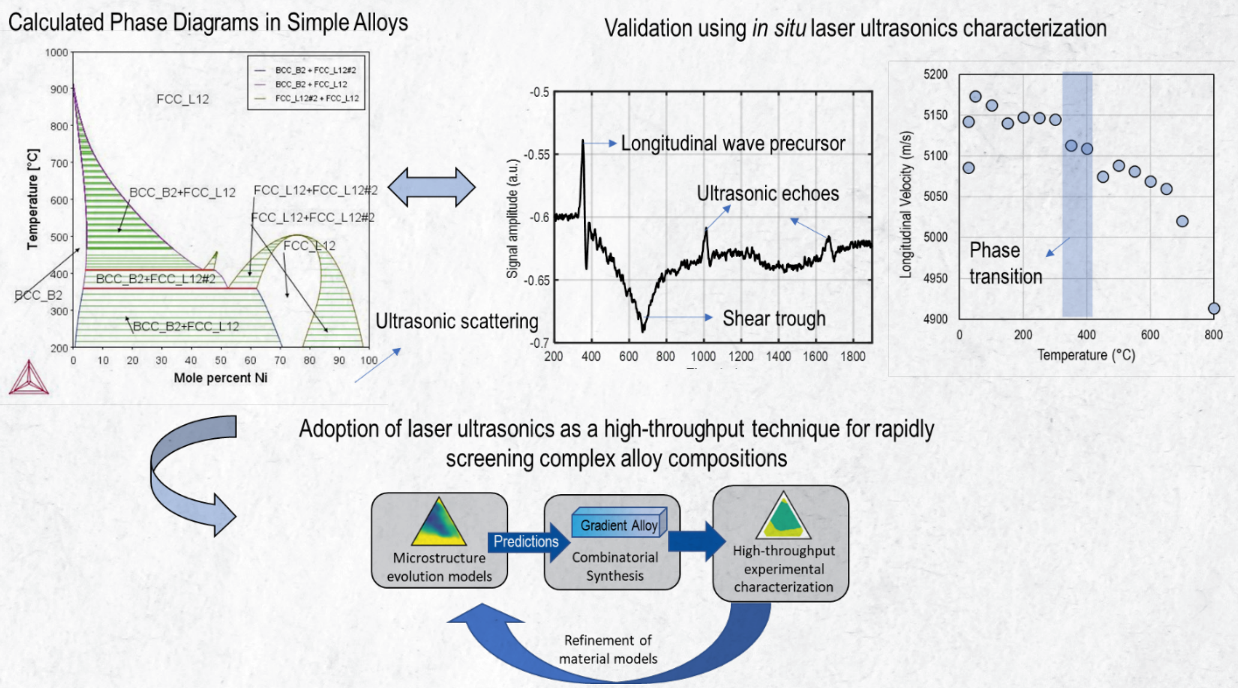
TOTAL APPROVED AMOUNT:
$110,000 over 1 year

PRINCIPAL INVESTIGATOR:
Amey Khanolkar

CO-INVESTIGATORS:
Austin Matthews, INL
David Hurley, INL
Dennis Tucker, INL
Subhashish Meher, INL
Wesley Jones, INL

Laser ultrasonics enables researchers to rapidly assess the temperature- and irradiation-induced phase transitions needed to develop novel materials for extreme environments.

This project utilized a laser ultrasonic technique to systematically study temperature-induced evolution of material properties in a set of interrelated binary alloys and a high entropy alloy fabricated using arc melting and spark plasma sintering processes. This technique involved the use of a nanosecond duration, high intensity pulsed laser to thermo-elastically generate ultrasonic waves that propagate in the bulk of the metal alloy. Sub-nanometer scale displacements associated with the propagating bulk ultrasonic waves were detected along the epicenter on the opposite surface of the sample using a 1 GHz bandwidth photorefractive interferometer. Phase transformations and microstructural changes were inferred from the temperature-dependent trends in the bulk acoustic velocities and features in the ultrasonic epicentral waveforms measured in the binary alloys. These inferences were then correlated with electron and optical microscopy observations and predictions using calculations of phase diagrams. The results in the set of binary alloy samples showed that the laser-generated ultrasonic pulses were strongly influenced by changes in material microstructure, accurately tracked thermally driven phase transformations, and detected the presence of microscale heterogeneities such as grain boundaries, dendritic structures, etc. The measurement approach was then applied to a quinary high entropy alloy sample to estimate phase transition temperature and determine microstructural heterogeneity. The rapid, non-contact and non-destructive ultrasonic testing approach demonstrated here is amenable to high throughput combinatorial investigations that can be applied to graded composition high entropy alloys produced using advanced manufacturing methods. When paired with atomistic simulations and calculations of phase diagrams modeling, this approach can overcome the bottlenecks faced by current material characterization methods to efficiently screen the vast discovery space of high entropy alloys that spans over a hundred million unique quinary alloy compositions.



This project developed an approach that used the laser ultrasonics technique for rapidly scanning structural phase transitions and microstructure evolution in metal alloys. This approach is non-contact, non-destructive, and is amenable to high throughput combinatorial investigations for efficiently screening the vast composition space of high entropy alloys.

TALENT PIPELINE:

- Amey Khanolkar, *Russell L. Heath Distinguished Postdoctoral Fellow*, converted to staff

PRESENTATION:

Khanolkar, A., S. Meher, D. H. Hurley, "Rapid Screening of High Entropy Alloys using Laser Ultrasonics," *Sixth International Workshop on Structural Materials for Innovative Nuclear Systems (SMINS-6)*, 12–15 Sep. 2022, Idaho Falls, ID, USA.

Feasibility Study of an Optical Fiber Instrument to Monitor Microstructure of Spark Plasma Sintering Products

PROJECT NUMBER:
21A1055-032

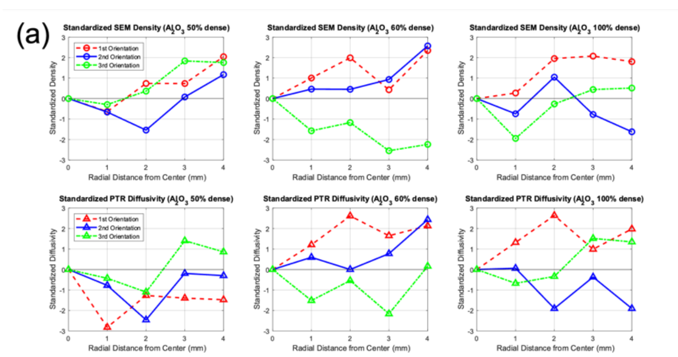
TOTAL APPROVED AMOUNT:
$125,000 over 1 year

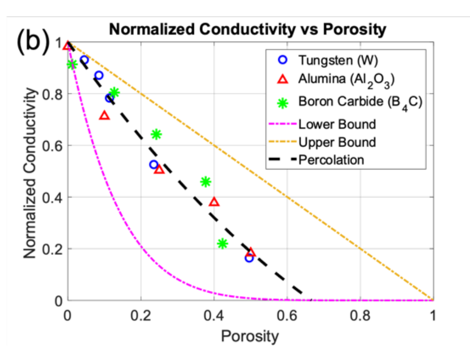
PRINCIPAL INVESTIGATOR:
Zilong Hua

CO-INVESTIGATORS:
Nathan Jerred, INL
Robert Schley, INL
Troy Holland, INL

Local thermal transport properties can be used to remotely examine microstructure features in spark plasma sintered products.

Spark plasma sintering is a rapid sintering technique that enables the densification and fabrication of a broad range of materials in an efficient way. High heating rates of hundreds of degrees Celsius per minute can be achieved, allowing materials to reach target temperatures in a short time. However, the large temperature gradients generate unexpected microstructure heterogeneities, degrading product mechanical, thermal, and electrical performance. Currently, no experimental approach exists to monitor such microstructural variation. In this project, the proof-of-concept methodology of using local thermal transport property to index the microstructure features, such as density variation, inside spark plasma sintering sintered products was investigated. A few series of materials were sintered using spark plasma sintering and their local thermal diffusivities were investigated using photothermal radiometry. We measured local thermal diffusivities along several radial directions across the samples, and the results qualitatively agree with the local porosities observed by using scanning electron microscopy. The analytical model that quantitatively correlates porosity to thermal property that can be used to estimate local porosity from measuring thermal diffusivity was further investigated. This study sets up the fundamental work to develop and deploy the photothermal radiometry-based system in spark plasma sintering to monitor the microstructure evolution during sintering. While thermal diffusivity can be measured in a real-time manner to estimate the bulk porosity of sintered materials without interrupting the sintering process, it can also examine local porosity variation so that the sintering parameters can be dynamically adjusted to enhance desired microstructure features, such as to fabricate functionally graded materials, or avoid unexpected heterogeneities.





(a) Qualitative comparison of results between local density characterized by using scanning electron microscopy and local thermal diffusivity measured by using photothermal radiometry on alumina samples; (b) quantitative relationship (the percolation model) of normalized thermal conductivity versus porosity with all samples.

Recovering Cobalt from Spent Lithium-ion Batteries Using Electrochemical Membrane Reactor without Chemical Consumption

PROJECT NUMBER:
21A1057-024

TOTAL APPROVED AMOUNT:
$135,000 over 1 year

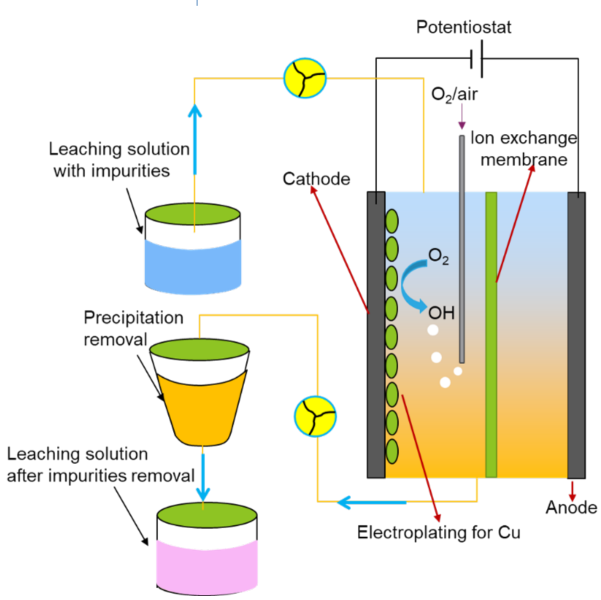
PRINCIPAL INVESTIGATOR:
Qiang Wang

CO-INVESTIGATORS:
Luis Diaz Aldana, INL
John Klaehn, INL

Novel electrochemical purification process removes impurities from spent lithium-ion batteries with no waste generation.

Expanding the electric vehicle market brings with it exponential growth in the use of lithium-ion batteries, which need lithium and transition metals including cobalt, nickel, and manganese for their cathodes. The import reliance of lithium, cobalt, nickel, and manganese in United States is 25–50%, 78%, 48%, and 100%, respectively. Recycling spent lithium-ion batteries is an effective strategy to mitigate the risk of supply chain disruption. Hydrometallurgy is the most promising methodology to recycle lithium-ion batteries because it is more efficient with better selectivity and less energy use. To recycle the spent batteries at an industrial scale, hydrometallurgy has the drawbacks of high chemical consumption, inducing high cost and negative environmental impact. In this project, a novel electrochemical membrane reactor was designed and proven. The reactor removed the impurities from lithium-ion batteries, including copper, iron and aluminum, to approximately 1 ppm by just using electricity, air, and water. The purified leachate maintains 99.5% of the nickel, 95.4% of the cobalt and 99.14% of manganese from the original leachate solution, and the leachate can be directly applied to cathode precursor synthesis. In comparison, the chemical precipitation method consumes a significant amount of chemicals, such as sodium hydroxide, and maintains only 55.2% of nickel and 76.1% of cobalt. Additionally, the electrochemical purification process does not introduce extra impurities, and the reactor restoration process generates valuable by-product sulfuric acid. This electrochemical process can reduce the cost occurred by material consumption due to the much less chemical consumption and the valuable by-product generation. The process mitigates waste emissions because no impurities are introduced, and no greenhouse gas is produced.

Diagram of electrochemical membrane reactor.



INTELLECTUAL PROPERTY:

Wang, Q., L. A. Diaz Aldana, D. M. Ginosar, M.Shi, "A Time-Efficient Electrochemical Membrane Reactor with Gas Diffusion Electrode as Cathode for Impurities Removal from Spent Li-ion batteries," Patent application date (Mar. 22, 2022).

Wang, Q., R. V. Fox, "A Flowsheet for Recovering all the Valuable Elements from Spent Lithium-ion Batteries without Waste Emission," (Sept. 12, 2022).

Development of Scalable Design Optimization Parameters for Bi-component Protective Systems

PROJECT NUMBER:
21P1056-004

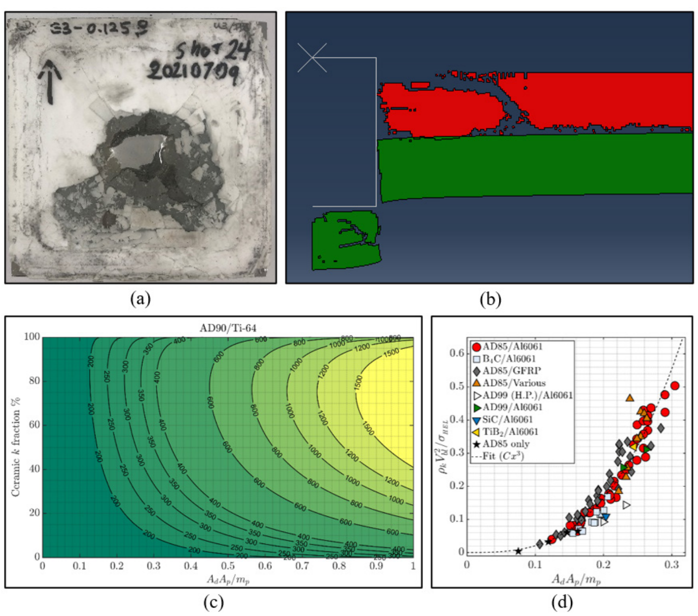
TOTAL APPROVED AMOUNT:
$254,000 over 2 years

PRINCIPAL INVESTIGATOR:
Zherui Guo
Russell L. Heath Distinguished Postdoctoral Fellow

CO-INVESTIGATOR:
Thomas Lillo, INL

Dimensionless scaling parameters predict performance metrics for rapidly deploying ceramic coated bi-component protective systems under extreme loading events.

Bi-component protection systems need to withstand a variety of extreme loading conditions such as high velocity projectile impact. Ceramics are widely used as the frontal material due to their high strength-to-weight ratios. This project identified the scaling laws linking material and geometric scales of ceramic coated bi-component light armor systems to the final impact performance. The impact failure dynamics for different projectile and target density ratios and relative component mass fractions were quantified and optimized for bonded alumina ceramic and titanium alloy systems. The mechanical wave and failure propagation within the target system was characterized and quantified via ballistic experiments, and experimental results were subsequently used to calibrate numerical simulations in Abaqus/Explicit. Using the respective wave propagation and solid mechanics equations for high velocity projectile impact, dimensionless parameters were derived for first-order performance predictions of armor systems and verified using existing data in literature. Different material classes were shown to have a characteristic strength term capable of collapsing the ballistic performance data. For ceramic coated armors, this characteristic term is the Hugoniot elastic limit, an effective yield strength for ceramic materials. The bonding strength between the components also affects the plate flexural stiffness of the overall structure and the shear stresses at the interface, both of which are major factors in determining the failure of the ceramic component. The contributions of inter-component bonding were parametrized based on the effective target stiffness and bending energy obtained from computational simulations.



a) Postmortem image of impacted alumina/titanium bi-component target; b) computational simulation of ballistic impact; c) optimization map of bi-component armor system; and d) collapsed ballistic perforation data using derived dimensionless parameters.

TALENT PIPELINE:

- Zherui Guo, *Russell L. Heath Distinguished Postdoctoral Fellow* converted to staff

PUBLICATION AND PRESENTATIONS:

Guo, Z., "Rate-dependent dynamic cylindrical cavity expansion equations for conical- and ogival-nosed projectiles," *Journal of Dynamic Behavior of Materials* (2022).

Guo, Z., "Dimensionless parameters to categorize the failure modes of ductile plate perforation." Proceedings of the 32nd International Ballistics Symposium, Reno, Nevada, USA (2022).

Guo, Z., "On Brittle Fracture Conoid Angles," *Bulletin of the International Ballistics Society*, (Projected publication Oct. 2022).

Water Soluble Support Material for Directed Energy Deposition Additive Manufacturing

PROJECT NUMBER:
21P1064-021

TOTAL APPROVED AMOUNT:
$123,000 over 1 year

PRINCIPAL INVESTIGATOR:
Joshua Zelina

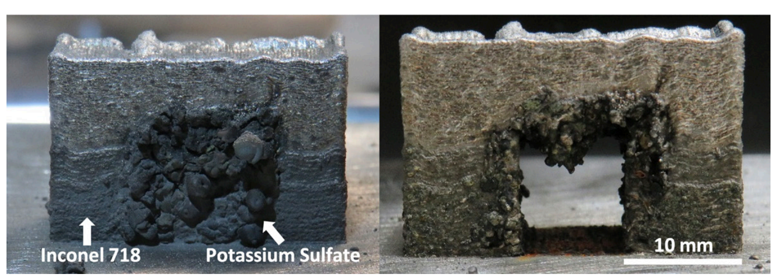
CO-INVESTIGATORS:
Fidelma Di Lemma, INL
Prabhat Tripathy, INL
Randall Scott, INL

INL scientists developed and demonstrated first known water soluble support material for metal additive manufacturing.

Metal additive manufacturing, specifically through directed energy deposition processes, offers an innovative alternative to traditional manufacturing. Additive manufacturing provides a method of fabricating complex geometries and highly customized three-dimensional parts that traditional manufacturing methods cannot achieve. However, metal additive manufacturing faces several key hurdles to fully realize its potential in industry. Among them, post processing issues are recognized as a significant challenge because current support material technology has a direct negative effect on post processing efforts. Support structures are made of the same metal as the target part and are thus essentially welded to the end part. Manual machining is required for support structure removal, increasing time, potential for part damage, and material waste in post processing while greatly decreasing design freedom. They are, however, necessary for printing complex geometries such as overhangs and internal features.

This project addressed this issue by developing a water soluble support material compatible with metal additive manufacturing. High melting temperature, water soluble salts were chosen for testing to fill this role. Potassium sulfate performed best and was printed alongside Inconel 718 as a support material. The Inconel 718 part featured a 90° bridge supported by the potassium sulfate. This feature would not be printable without a support material, and thus provided the simplest test case to prove potassium sulfate's viability as a support material. The multi-material part printed successfully. The Inconel 718 and potassium sulfate part was then placed into deionized water that dissolved the potassium sulfate supports and left behind the Inconel 718 structure. This demonstrated the first known water soluble support material compatible with metal additive manufacturing, which represents a significant advancement in the additive manufacturing field.

The development of a water soluble support material will enable the fabrication of next generation fuel designs that were un-manufacturable. Additionally, the project was the first to successfully demonstrate multi-material directed energy deposition additive manufacturing at INL, which will be instrumental in informing future research initiatives. Finally, this research demonstrated the first known directed energy deposition additive manufacturing of salts. In summary, the project achieved its goals and yielded a novel additive manufacturing capability for use in INL and industry.



Additively manufactured Inconel 718 part with 90° overhang printed with potassium sulfate support structure. Shown before dissolving support structure (left) and with support structure dissolved (right).

SECURE AND RESILIENT CYBER-PHYSICAL SYSTEMS

Relying on its world-class research and development capabilities and unique assets, INL secures our nation's critical civilian infrastructure and military systems against complex and dynamic cyber threats while increasing resiliency to a variety of hazards and environmental changes. INL advances transformational approaches addressing true risks throughout critical system lifecycles. INL's core capabilities facilitate efforts to formalize and scale cyber-informed science and engineering, strengthen all-hazards physical and cyber critical infrastructure resilience, innovate for enduring control systems cybersecurity, establish a secure resilient digital supply chain for critical infrastructure, and secure wireless communications and spectrum use as a key enabler to critical systems.

Resilient Attack Interceptor for Intelligent Devices

PROJECT NUMBER:
20A44-003

TOTAL APPROVED AMOUNT:
$660,000 over 3 years

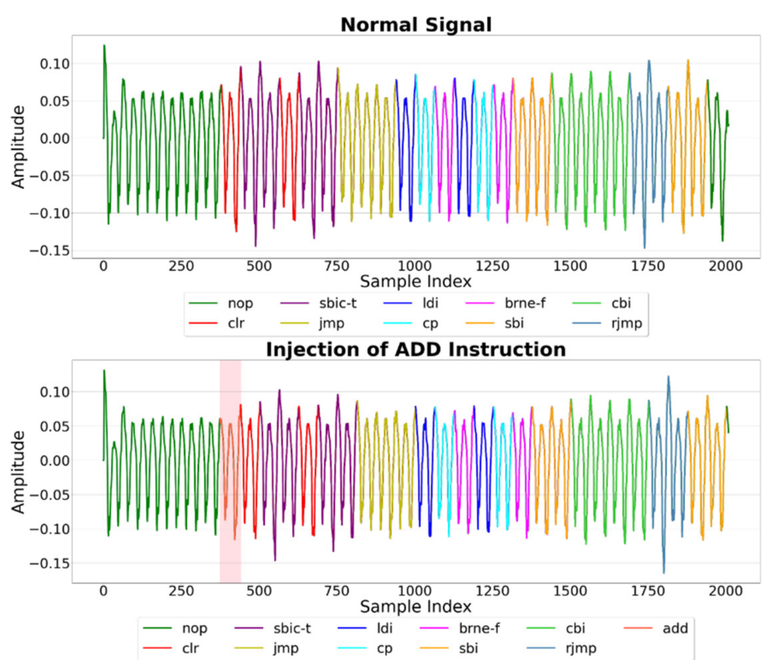
PRINCIPAL INVESTIGATOR:
Craig Rieger

CO-INVESTIGATORS:
Jacob Ulrich, INL
Robert Ivans, INL
Costas Kolias, University of Idaho

COLLABORATOR:
Cynalytica

Detecting nefarious control system exploitation by recognizing and responding to distributed side channel anomalies ensures critical control system operations.

INL and the University of Idaho Resilient Attack Interceptor for Intelligent Devices developed external monitoring methods to protect operational technology and industrial Internet of Things devices by collecting and analyzing observable physical aspects that are produced naturally and involuntarily during the operational life cycle with anomalous functionality. More specifically, analog signals—physical side channels—that may be used for this purpose are electromagnetic emissions, power consumption, thermal profile, or acoustic activity of a protected device and its components. The developed system relied on two side channels—electromagnetic and thermal signals along with conventional network traces and the collected observations—fused using a novel approach that integrates scalable physics relationships with unsupervised anomaly detection methods. The developed technology provided an analytical approach to recognize several different attack types that include denial of service, data injection, and others over a large set of industrial Internet of Things device types. A primary design tenet was the decoupling of the proposed monitoring system from the protected system and its normal operation. The resulting patent pending technology provides an alert with physical or plant system context for disposition, allowing for human-in-the-loop orchestration and potentially automated response, and is currently being marketed for commercialization.



The solution developed under this research can detect the injection of a single instruction at the assembly level (bottom).

TALENT PIPELINE:

- Robert Ivans, graduate fellow at INL, converted to staff
- Georgios Michail Makrakis, student at University of Idaho
- Haotian Wang, student at University of Idaho
- Kurt Vedros, student at University of Idaho
- Ekaterina Miller, student at University of Idaho
- Alexander Hearn, high school student in Idaho Falls

PUBLICATIONS AND PRESENTATIONS:

Miller, E., G. M. Makrakis, K. A. Vedros, C. Kolias, C. Rieger, D. Barbara, "Detecting Code Injections in Noisy Environments Through EM Signal Analysis and SVD Denoising," *ESCS 21*, 2022.

Rieger, C., K. Schultz, T. Carroll, T. McJunkin, "Resilient Control Systems—Basis, Benchmarking and Benefit," *Institute of Electrical and Electronics Engineers Access* 9 (2021) 57565–57577.

Makrakis, G. M., C. Kolias, G. Kambourakis, C. Rieger, J. Benjamin, "Industrial and Critical Infrastructure Security: Technical Analysis of Real-Life Security Incidents," *Institute of Electrical and Electronics Engineers Access* 9, (2021) 165295–165325.

Vedros, K., G. M. Makrakis, C. Kolias, M. Xian, D. Barbara and C. Rieger, "On the Limits of EM Based Detection of Control Logic Injection Attacks in Noisy Environments," *2021 Resilience Week (RWS)* (2021) 1–9.

Kolias, C., D. Barbara, C. Rieger, J. Ulrich, "EM Fingerprints: Toward Identifying Unauthorized Hardware Substitutions in the Supply Chain Jungle," *CReSCT Workshop Institute of Electrical and Electronics Engineers Symposium on Security and Privacy*, May 2020.

Ivans, R., G. Makrakis, A. Hearn, K. Vedros, C. Kolias, C. Rieger, "Parallel Learning Machines for Complementary Thermal and Electromagnetic Side Channel Anomaly Recognition," submitted to *ACSAC Eighth Annual Industrial Control System Security Workshop*, Dec. 2022.

INTELLECTUAL PROPERTY:

Rieger, C. G., R. C. Ivans, C. Kolias, D. Barbara, "EM Based Detection of Control Logic Injection Attacks in Noisy Environments," Patent application (Oct. 15, 2021).

License 21-LA-69, "Scalable, Physical Effects Measurable Microgrid for Cyber Resilience Analysis," (Feb. 23, 2021).

Protocol Analytics to Enable Forensics of Industrial Control Systems

PROJECT NUMBER:
20A44-032

TOTAL APPROVED AMOUNT:
$1,157,000 over 3 years

PRINCIPAL INVESTIGATOR:
Keith Mecham

CO-INVESTIGATORS:
Daniel Hearn, INL
Devin Vollmer, INL
Tanmay Bhagwat, INL
Ted Tracy, INL

New methods to capture, identify, and translate traffic from any industrial control system network regardless of communication protocol expand the effectiveness of existing cybersecurity tools.

Cybersecurity tools available today are effective at monitoring traffic within information technology and operational technology networks standardized around Ethernet communication. However, although considered part of the operational technology network, many embedded and industrial control systems utilize proprietary or legacy communication protocols or both that are not compatible with Ethernet. This complicates efforts to secure operational technology networks against cyberattack because existing tools are unable to monitor or interface with devices that are in direct control of machinery, equipment, and processes. This visibility gap affords attackers an opportunity to operate within industrial control systems networks undetected until they cause an observable change or disruption to systems.

The primary objective of this research project was to eliminate the industrial control systems visibility gap by capturing traffic from the various industrial control systems networks and proving the feasibility of a universal industrial control systems translator. Researchers analyzed 13 key industrial control systems protocols representing the diverse spectrum of electrical and signal encoding attributes commonly used within United States critical infrastructure. Identifying electrical signatures were developed along with algorithms to identify target signals as they were acquired. Once successfully identified, researchers developed decode algorithms for each protocol capable of determining the data represented by the acquired signal.

This is noteworthy because the decode algorithm was realized entirely in software, and researchers eliminated hardware transceivers from the signal digitization path completely. Although hardware transceivers are ubiquitous across devices employing serial and Ethernet based communication, eliminating the need for dedicated hardware is a key prerequisite to a truly universal communication device. The next step was the development of a hardware prototype capable of capturing all the target communication signals, executing the developed identification and decode algorithms in real-time, and re-transmitting the captured data over Ethernet to an existing cybersecurity sensor/aggregator.

Initial bench-top testing and evaluation within simulated industrial control systems environments prove that the methods, algorithms, and circuitry that researchers designed support the development of a scaled-up device capable of capturing traffic from any industrial control systems network. Currently, the technology is patent pending, is in the process of being licensed for commercialization as "OmniTap," and will lead to enhanced protection of control systems within critical infrastructure.

TALENT PIPELINE:

- Conner Buck, student at Arizona State University
- Justin Leiden, student at Mississippi State University
- Trevor Smith, student at Boise State University

PRESENTATION:

Mecham, K., "OmniTap: Universal Capture and Translation of ICS Communication," *TechConnect World Innovation Conference and Exposition 2022*, National Harbor, MD, USA.

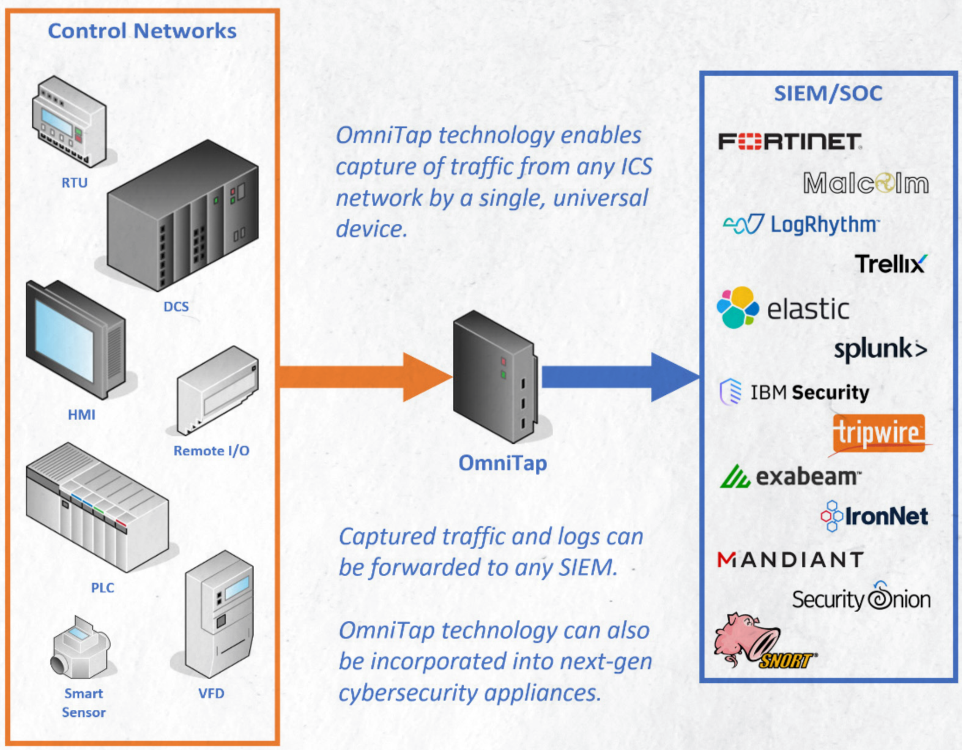
INTELLECTUAL PROPERTY:

Mecham, K. D., T. R. Tracy, D. P. Hearn, D. J. Vollmer, T. S. Bhagwat, "Universal Capture and Translation of Industrial Control System (ICS) Communication," Patent application (Nov. 17, 2021).

License 22-LOA-07, "Signals in a network," May 5, 2022.

AWARD:

TechConnect National Innovation Awardee – 2022



The OmniTap technology enables a universal device to monitor any industrial control systems network and will provide visibility to most control devices for the first time.

Automated Infrastructure and Dependency Detection via Satellite Imagery and Dependency Profiles

PROJECT NUMBER:
20A44-195

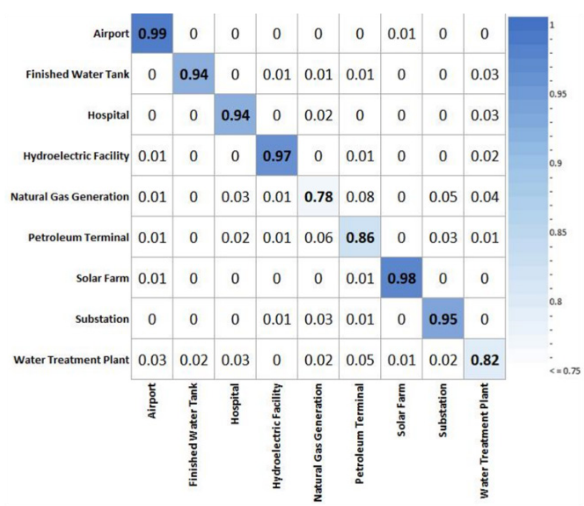
TOTAL APPROVED AMOUNT:
$797,500 over 3 years

PRINCIPAL INVESTIGATOR:
Shiloh Elliott

CO-INVESTIGATORS:
Ryan Hruska, INL
Iris Tien, Georgia Institute of Technology

In a first-of-class computer vision approach, explainable convolution neural networks identify critical infrastructure facilities.

This project had three main objectives: 1) establish if convolutional neural networks, a supervised machine learning modeling class, could correctly classify multiple critical infrastructure facilities in satellite images, 2) determine through the utilization of an explanability framework that the developed model demonstrated true accuracy, and 3) determine if it is possible to correctly classify the subcomponents of the identified critical infrastructure facilities, such as a transformer at a substation. To execute on these objectives, the research team established a modeling pipeline that used United States Department of Agriculture's National Agriculture Imagery Program as input data. This data set was chosen for the following reasons: it has relatively high resolution of 1 – 0.5 m, a data refresh rate of three years, coverage of the contiguous United States, and open-source accessibility. Through extensive experimentation, the research team created an explainable modeling pipeline capable of identifying nine critical infrastructure facilities with an average accuracy of 90% across classes. The explainability portion of the pipeline allows for confidence in the model's conclusion by determining which portions of the image were used in the classification activity. Objective three proved to be challenging. The research team established individual faster region-based convolutional neural network for four infrastructure classes. Results from these models varied widely with average precision—a proxy metric for accuracy—ranging from 10% to 70%. Ultimately, the project resulted in an explainable modeling pipeline capable of identifying the nine infrastructure classes of interest. This work can be expanded to include more infrastructure classes or retrained for another domain with possible applications in a range of national security areas from critical infrastructure research to counterintelligence.



A confusion matrix of accuracy for objective one.

TALENT PIPELINE:

- Ashley Shields, student at Idaho State University, converted to staff
- Danielle Barna, student at Colorado School of Mines
- David Finck, student at University of California Irvine
- Elizabeth Klaehn, student at Texas Lutheran University
- Keelie Lawler, student at Montana Technological University
- Shanon Brailsford, student at Idaho State University
- Tyler Paladino, student at Idaho State University
- Chase Christen, student at Boise State University

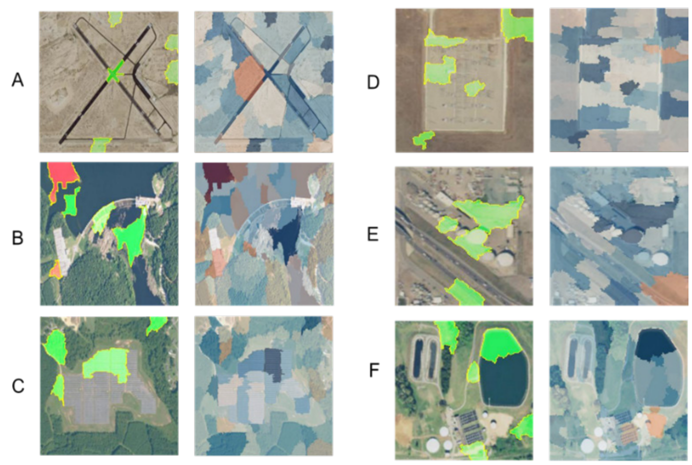
PRESENTATIONS:

Elliott, S., R. Hruska, A. Shields, "Utilizing Explainable Convolutional Neural Networks for Critical Infrastructure Analysis and Identification," *Military Operations Research Society 89th Symposium*, 2021.

Elliott, S., A. Shields, E. Klaehn, I. Tien, "Identifying Critical Infrastructure in Imagery Data Using Explainable Convolutional Neural Networks," *Remote Sensing*, 14(21) (2022) 5531.

INTELLECTUAL PROPERTY:

License 22-LA-42, "Scramble," Mar. 9, 2022.



Results of explainability for different critical infrastructure classes.

Advanced Machine Learning-based Fifth-Generation Network Attack Detection System

PROJECT NUMBER:
21A1050-026

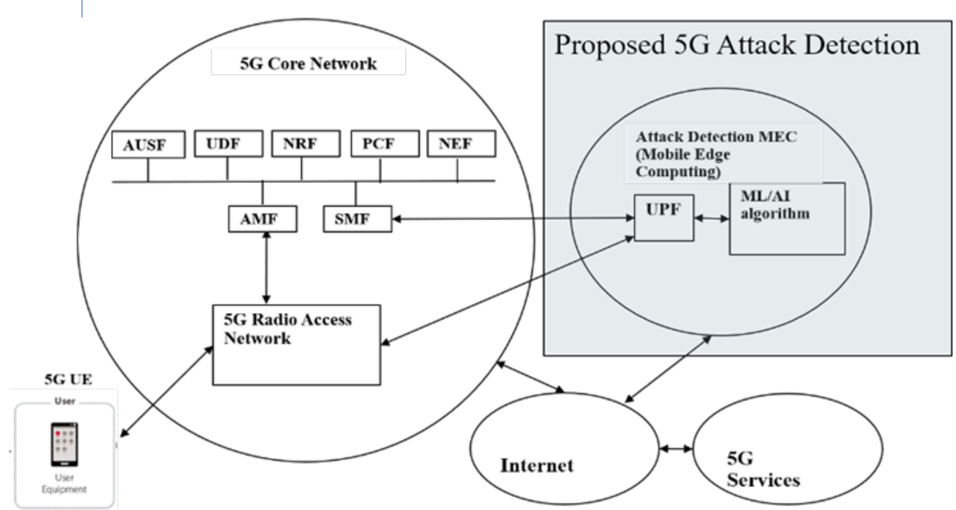
TOTAL APPROVED AMOUNT:
$725,000 over 2 years

PRINCIPAL INVESTIGATOR:
Jared Wadsworth

CO-INVESTIGATORS:
Kurt Durr, INL
Shad Staples, INL

Novel machine learning algorithm increases the detection accuracy of malicious traffic on encrypted 5G networks.

The goal of this project was to prove the principle that network attacks can be detected on a fifth-generation (5G) network using machine learning. To achieve this goal, we created multiple novel data sets that contained network traffic data alongside multiple types of malicious traffic. One of these data sets was created on a traditional network to simulate modern attacks, which is missing in most academic data sets. The other data set was created on a 5G network and includes novel attacks on the 5G core. In conjunction with these efforts, we tested the common academic approaches of network traffic analysis on the most used academic network packet data sets. We then applied multiple techniques from other fields of machine learning and artificial intelligence, including computer vision and natural language processing, to see if any of those methods could be applied in our setting. After testing many different structures of models, we determined that variational autoencoders provided the best results. During this analysis, we discovered the imbalanced nature of the data we and others have collected. Many of the common metrics are skewed toward lab environments and do not perform as well on live networks. To combat this issue, we created a generative adversarial network capable of mimicking both normal and malicious traffic. This allowed us to train our models on synthetic data in addition to real data, which allowed our models to train longer without biasing the results.



Architecture for detecting attacks at the edge of the 5G network via a machine learning / artificial intelligence algorithm.

TALENT PIPELINE:

- Justin Hales, student at Brigham Young University
- Hannah Cartier, student at University of Utah
- Jacob Rhodes, student at Kennesaw State University

PUBLICATIONS:

Wadsworth, J., J. Hales, S. Staples, "An Overview of the Usefulness of Machine Learning Techniques on Network Packet Data," *Institute of Electrical and Electronics Engineers Access* (n.d.).

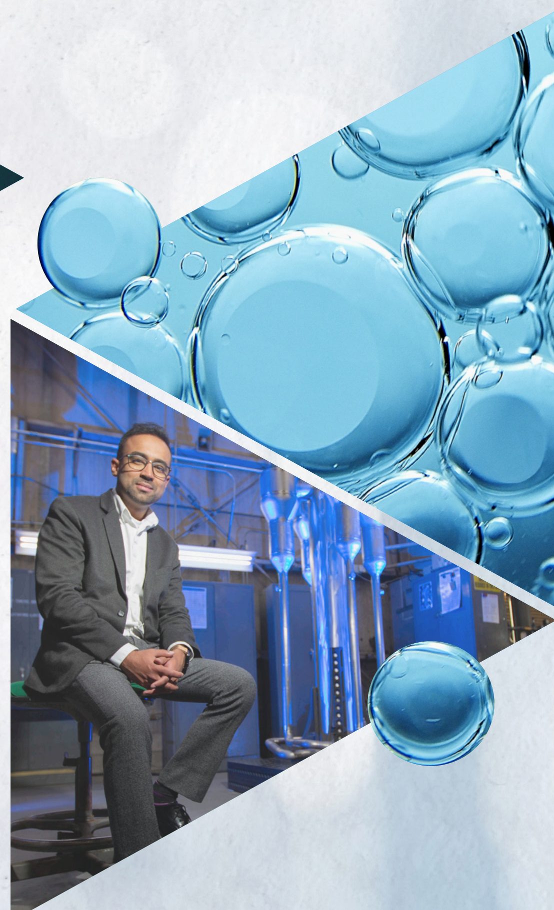
Wadsworth, J., H. Cartier, J. Rhodes, S. Staples, "A Representative Enterprise Environment Network Data set for Machine Learning," *Institute of Electrical and Electronics Engineers Access* (n.d.).

Wadsworth, J., H. Cartier, S. Staples, "Detecting malicious network traffic using variational autoencoders," *Institute of Electrical and Electronics Engineers Access* (n.d.).

Wadsworth, J., S. Staples, "Increasing the Effectiveness of Network Packet Models using Generative Adversarial Networks," *Institute of Electrical and Electronics Engineers Access* (2022).

"The art and science of asking questions is the source of all knowledge."

– Thomas Berger, American Novelist



Battelle Energy Alliance manages INL for the U.S. Department of Energy's Office of Nuclear Energy

Idaho National Laboratory