INL/RPT-22-66730-Revision-0

2021 Annual Report Laboratory Directed Research & Development

April 2022

Changing the World's Energy Future

Tony Huff, Allison Lee Hummel, Stephannie A Lambert

Idaho National Laboratory

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

INL/RPT-22-66730-Revision-0

2021 Annual Report Laboratory Directed Research & Development

Tony Huff, Allison Lee Hummel, Stephannie A Lambert

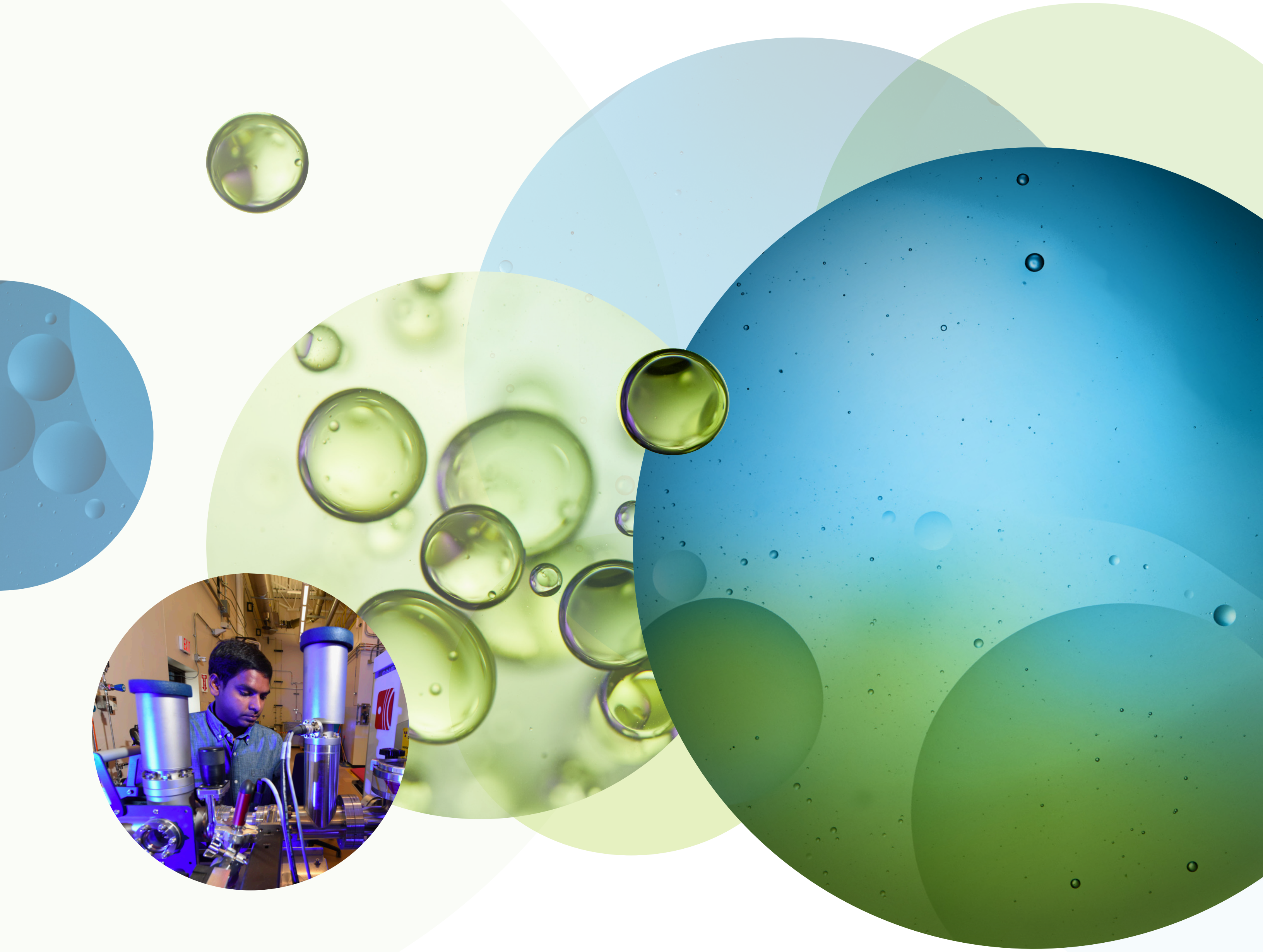
April 2022

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-22-66730



2021
ANNUAL REPORT
Laboratory Directed Research & Development

Idaho National Laboratory

This report provides an overview of the main accomplishments of the LDRD program in FY21.

LETTER FROM INL'S CHIEF RESEARCH OFFICER

The Department of Energy's (DOE) Laboratory Directed Research and Development (LDRD) program is an essential pathway for innovation, capability growth, and research staff development at Idaho National Laboratory (INL). This program enables timely and agile response to national security, energy, and environmental challenges that motivate INL's mission to discover and demonstrate innovative nuclear energy solutions and other clean energy options as well as securing our critical infrastructure. This report highlights INL's LDRD projects concluding in fiscal year (FY) 2021 which included innovative research and development (R&D) across INL's five science and technology initiatives: nuclear reactor sustainment and expanded deployment, integrated fuel cycle solutions, integrated energy systems, advanced design and manufacturing for extreme environments, and secure and resilient cyber-physical systems.

INL's LDRD portfolio grew 60% over the last five years from $23 million in FY17 to $37 million in FY21. In FY17, 84 projects were supported as compared to 147 projects in FY21. Our program employs a rigorous review and selection process to identify the most innovative research projects that support INL's mission and provide the greatest impact to our sponsors. The increased number of projects reflects an increased number of high-quality proposals, which, in turn, indicates increasingly innovative thinking by INL researchers.

Dr. Marianne Walck

DEPUTY LABORATORY DIRECTOR
FOR SCIENCE AND TECHNOLOGY
AND CHIEF RESEARCH OFFICER
IDAHO NATIONAL LABORATORY

From FY17 to FY21, the 60% increase in budget with the 75% increase in the number of supported projects also indicates the success of the Seed LDRD program. First piloted in FY18 and fully established in FY20, Seed projects are limited to 12 months duration and $125k funding. The Seed program funds innovative ideas and enables researchers to test high-risk hypotheses. The program allows researchers to explore domains beyond the five science and technology initiatives while remaining aligned with INL's mission.

INL's distinguished postdoctoral fellowship program continues to grow and attract scientists embarking on their research careers. The LDRD program supports approximately half of the distinguished postdocs' time through projects for which they are the principal investigators. In FY21, the six distinguished postdoc proposals were approved for funding with a total LDRD project budget exceeding $1.3M.

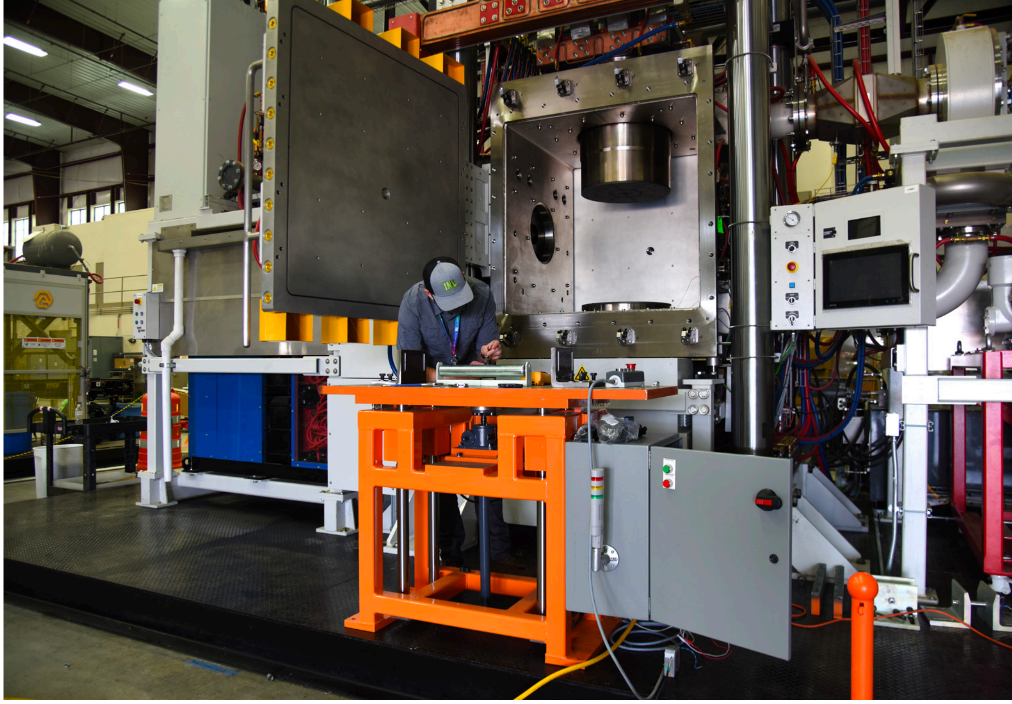
Every five years, DOE publishes an LDRD highlights brochure that showcases cutting-edge research projects from the LDRD programs at all DOE laboratories. In FY21 alone, there were 2,394 projects according to DOE's FY21 LDRD Report. To be included in DOE's highlights brochure, therefore, is an impressive honor. INL's project "Production of Scandium-47 for Potential Use as a Radiotheragnostic," led by principal investigator Dr. Mathew Snow and featured on page 64 in this report, received this honor. This project exemplifies the LDRD program intent – innovative impactful research, collaborative capability growth, and scientific staff development.

INL researchers are submitting more high-quality, high-value proposals than ever before and more than the current budget allows us to fund. We plan to continue growing INL's LDRD program and enabling our world-class staff to address DOE's current and future missions with leading-edge research. I invite you to review this report to see how INL's LDRD portfolio enables innovation and builds researcher talent to advance INL's vision to change the world's energy future and secure our critical infrastructure.

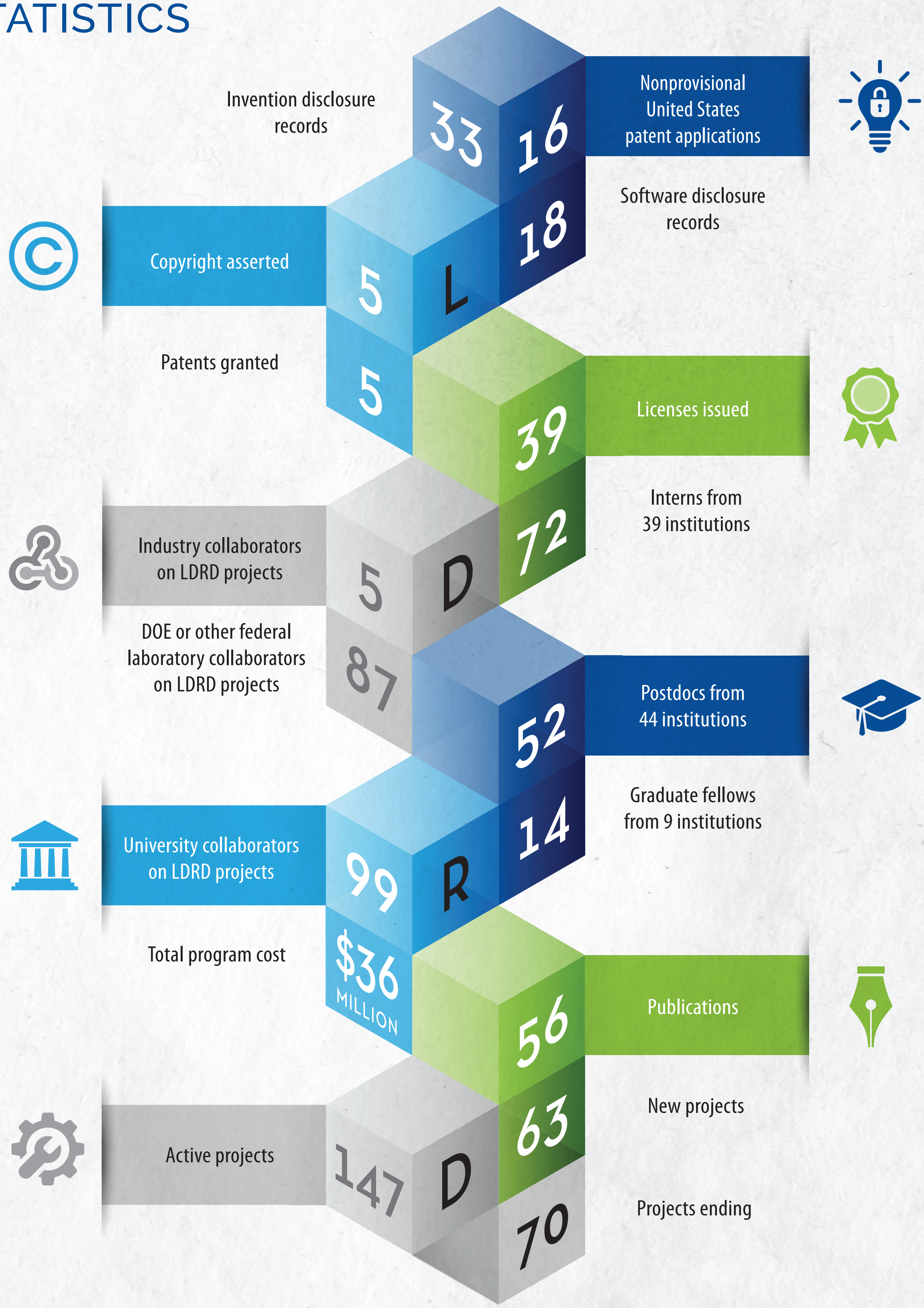
TABLE OF CONTENTS

Integrated Fuel Cycle Solutions 54

Integrated Energy Systems 73

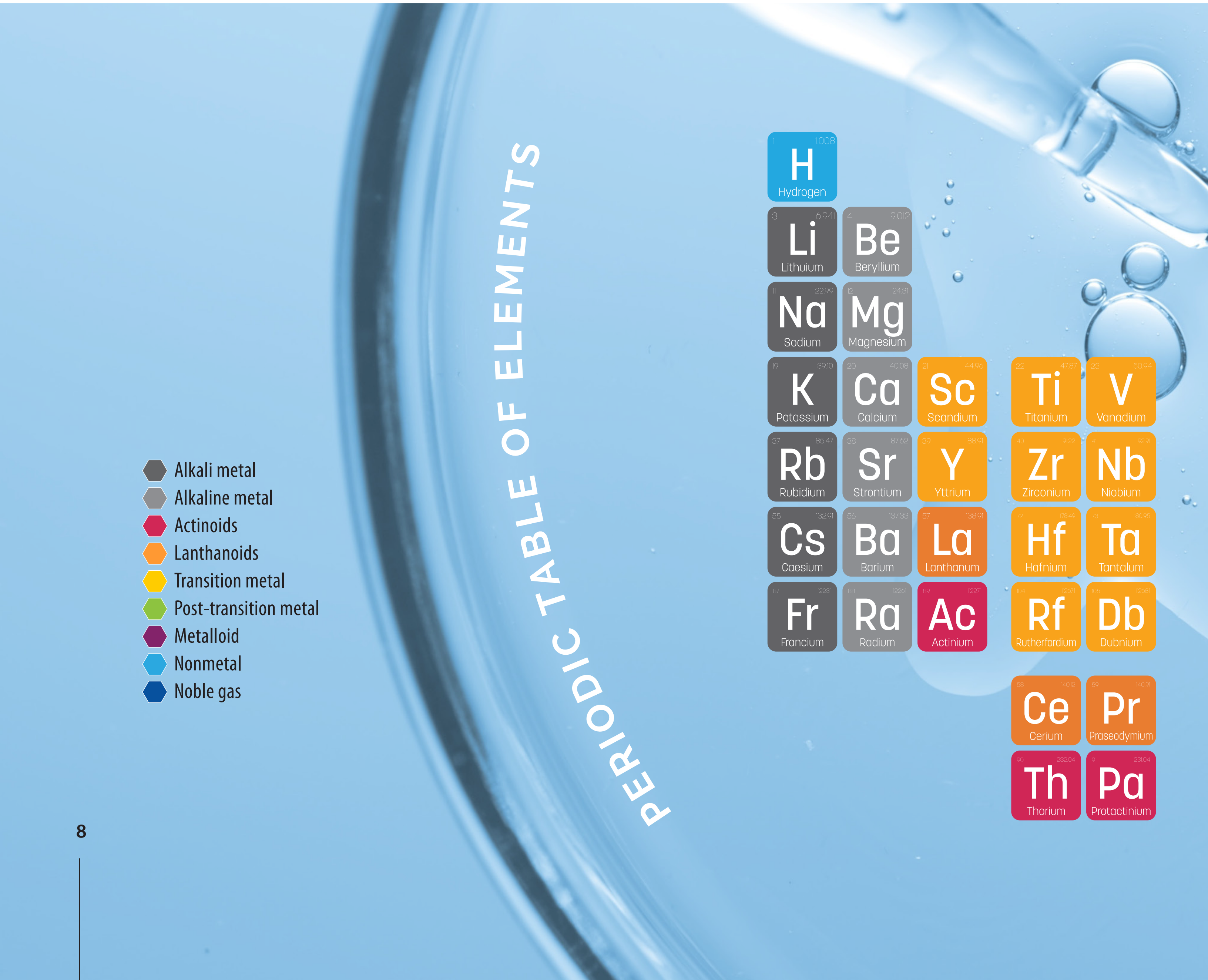


FY 2021 PROGRAM STATISTICS

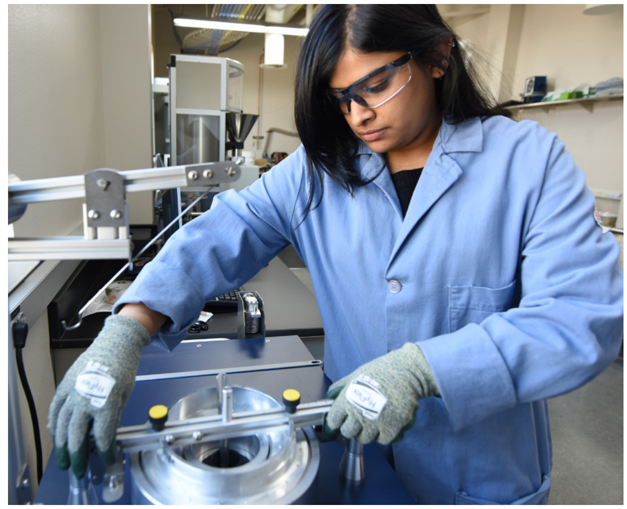


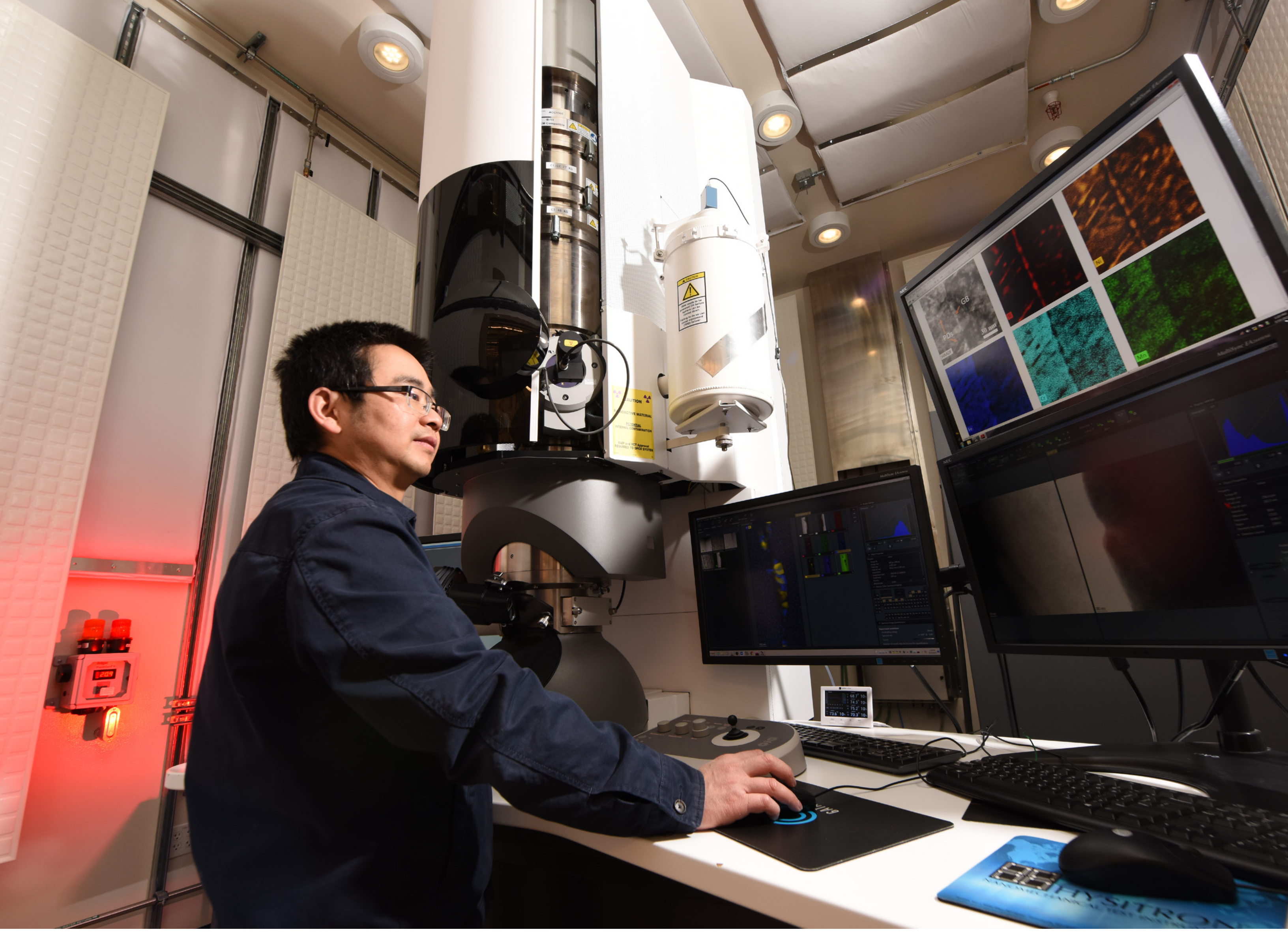
ACRONYMS & ABBREVIATIONS

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





LDRD OVERVIEW

INL's LDRD Portfolio

The diverse LDRD projects explore a range of scientific and engineering concepts at the forefront of nuclear energy, national and homeland security, and energy and environment. INL's LDRD portfolio comprises four investment components that are continually aligned with INL's vision, mission, and science and technology initiatives.

- The strategic R&D fund supports research that advances INL's science and technology initiatives.
- The seed fund supports high impact, innovative research that is aligned with INL's mission, even if not explicitly aligned with a science and technology initiative.
- The distinguished postdoc fund supports early career researchers in INL's three distinguished postdoctoral fellowships, providing them leadership opportunities while they conduct leading edge research that supports INL's mission.
- The strategic hire fund supports mid to late career researchers, providing opportunities to advance their scientific leadership while advancing INL's mission.

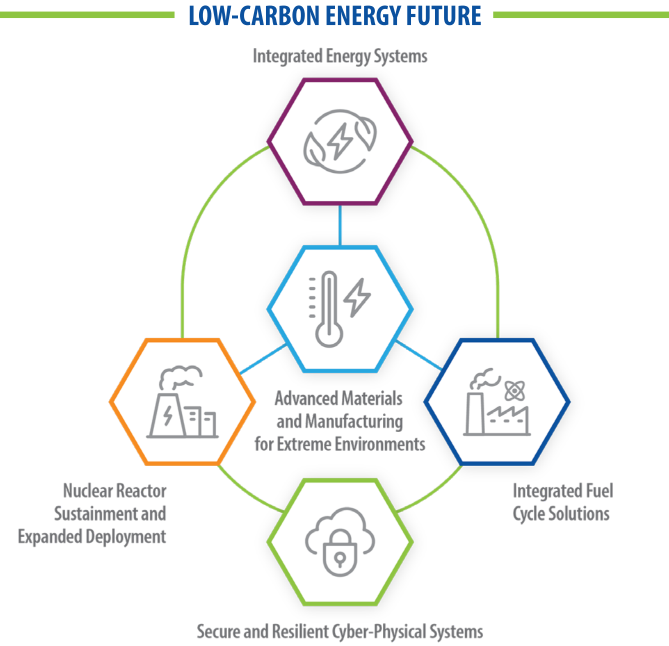
Core Capabilities

Of the 24 core capabilities identified by DOE as foundational to its mission, DOE acknowledges that INL has 13 core and two 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 R&D.

These core capabilities are sustained and enhanced through INL's LDRD projects. To begin demonstrating that the emerging core capabilities are intrinsic at INL, the LDRD program added the two emerging core capabilities—Chemical and Molecular Science and Condensed Matter Physics and Materials Science—as strategic scientific initiatives. Six projects supporting INL's mission were approved for funding in these two areas.

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 project 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.

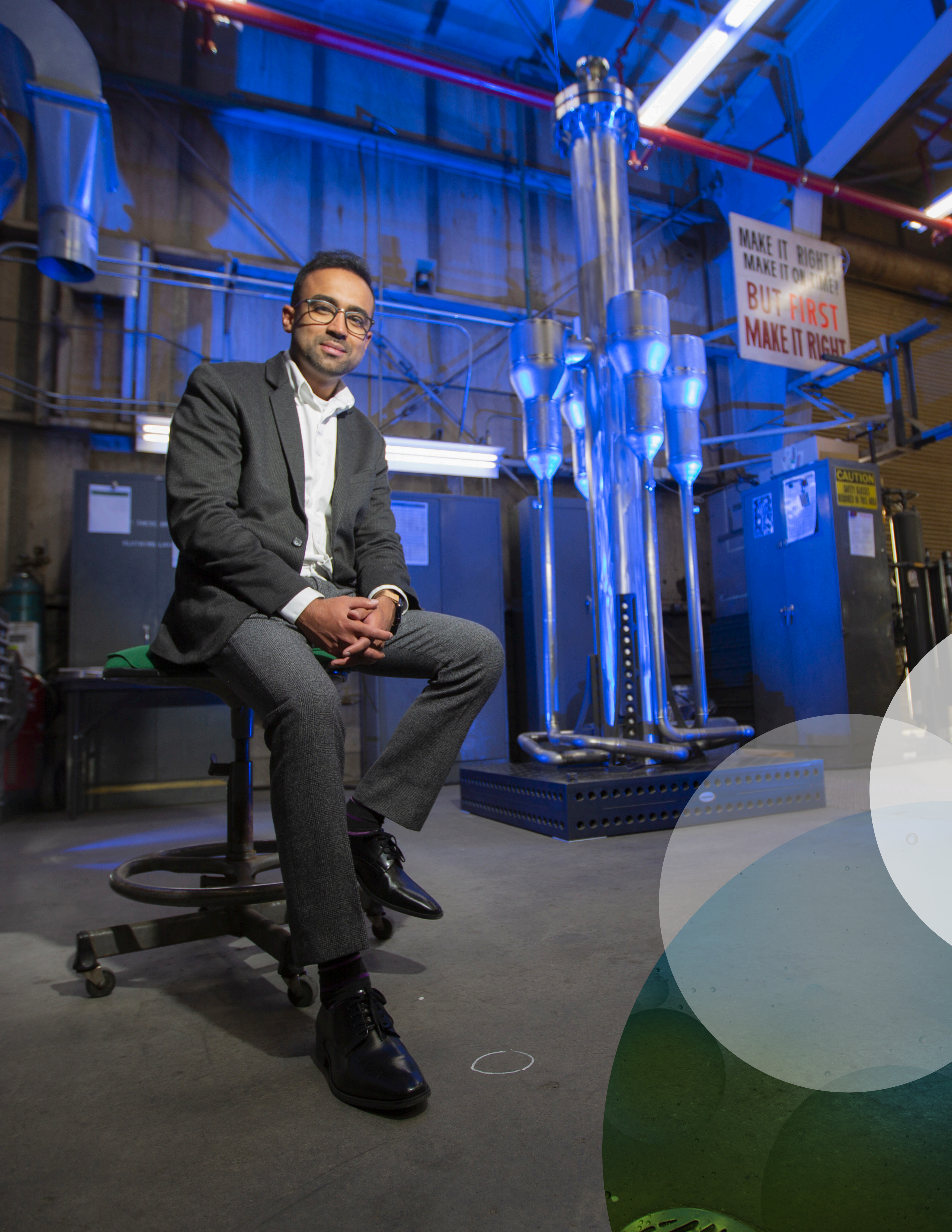


- Advanced computer science, visualization, and data
- Applied materials science and engineering
- Biological and bioprocess engineering
- Chemical and molecular science (emerging)
- Chemical engineering
- Condensed matter physics and materials science (emerging)
- Cyber and information sciences
- Decision science and analysis
- Environmental subsurface science
- Large-scale user facilities/R&D facilities/ advanced instrumentation
- Mechanical design and engineering
- Nuclear and radiochemistry
- Nuclear engineering
- Power systems and electrical engineering and integration
- Systems engineering and integration

FOUNDATIONAL CORE CAPABILITIES

Showcasing Success

On September 9, 2021, INL hosted a virtual poster session showcasing the LDRD projects ending in FY21. The virtual poster session mimicked traditional poster session interactions by facilitating chat access to principal and co-investigators and by providing all principal investigators with a presentation slot. Attendees included INL researchers, industry and academic partners, external collaborators, and members of the public.


MAKE IT RIGHT
BUT FIRST
MAKE IT RIGHT
CAUTION

NUCLEAR REACTOR SUSTAINMENT AND EXPANDED DEPLOYMENT

The clean, reliable energy produced by commercial nuclear reactors is essential to a net-zero energy future. Today, nuclear energy provides nearly 52%[1] of the carbon-free electricity in the United States, more than any other source. In the near future, nuclear plants will complement wind, solar, fossil fuels with carbon capture and storage, and innovative energy storage options to make net-zero achievable. INL leads innovative nuclear research, development, and demonstration (RD&D) to meet the challenge of increasing net-zero energy options in the face of growing energy needs by sustaining and extending the safe and economical operational life of the current commercial fleet while concurrently working to deploy a new generation of advanced. INL leverages its strategic infrastructure and strategic partnerships with industry and other stakeholders to create and define the next phase of global nuclear energy by driving technological innovations and operational advances through proof of concept, proof of performance, and proof of operation.

1 https://www.energy.gov/ne/articles/5-fast-facts-about-nuclear-energy

Real-time Transient Heating Measurements for Advanced Experiment Design and Model Validation

TOTAL APPROVED AMOUNT:
$723,651 over 3 years

PROJECT NUMBER:
18A12-020

PRINCIPAL INVESTIGATOR:
Nicolas Woolstenhulme

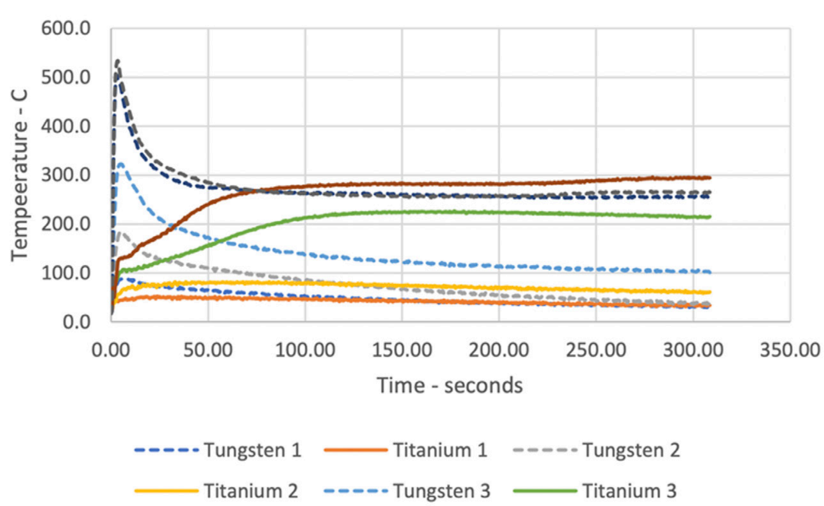
CO-INVESTIGATORS:
Colby Jensen, INL
James Parry, INL
Joshua Daw, INL
Lance Hone, INL

Demonstrated proof-of-principle blazes trail for ultrasonic thermometer type instruments to measure time-dependent nuclear heating in transient reactor environments.

Development, demonstration, and licensing of nuclear fuels requires irradiation testing of specimens. The magnitude of nuclear heating is one of the most crucial attributes in irradiation test design, analysis, and data interpretation. This project demonstrated a novel nuclear heating sensor technology that directly measures this crucial parameter using spatially resolved real-time ultrasonic thermometry. This sensor was demonstrated by irradiation in the Transient Reactor Test facility (TREAT). This instrument measures nuclear heating via temperature-based calorimetry made in real-time based on acoustic time-of-flight shift arising from temperature dependent changes the material properties of nuclear-heated wires. The proof-of-principle for this sensor was performed with an electrically heated waveguide wire arranged as a classical ultrasonic thermometer. Since the sensor was used to measure self-heating, rather than environmental temperature, it was referred to as an ultrasonic thermometer calorimeter. Successful construction and irradiation of the ultrasonic thermometer calorimeter was performed on a non-uranium version using prompt gamma heating to heat the waveguide wire.

PRESENTATION:

Fathi, N., P. McDaniel, N. Woolstenhulme, L. Hone, J. Daw, E. Larsen, K. Tsai, J. Parry, and C. Jensen, "Evaluation of a self-heated ultrasonic calorimeter for in-reactor application," *International Conference Nuclear Energy for New Europe*, 2020.



Example ultrasonic thermometer calorimeter data during irradiation where the prompt temperature rise can be seen in the first few seconds.

In-pile Investigation of Transient Boiling in the Transient Reactor Test Facility

TOTAL APPROVED AMOUNT:
$2,650,000 over 3 years

PROJECT NUMBER:
18P37-001

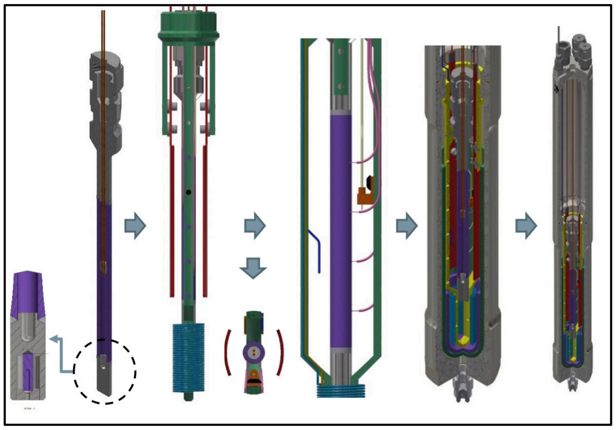
PRINCIPAL INVESTIGATOR:
Colby Jensen

CO-INVESTIGATORS:
Nicolas Woolstenhulme, INL
Nicholas Brown,
University of Tennessee, Knoxville
Richard Christensen,
University of Idaho

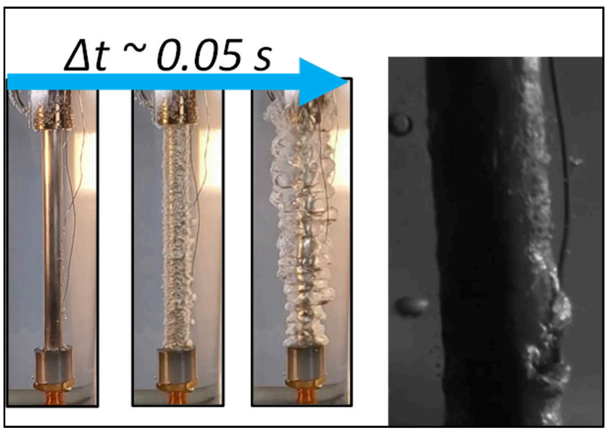
Improved cladding-to-coolant heat transfer understanding can increase safety and design margins in both the current commercial fleet and advanced light water reactors.

Cladding-to-coolant heat transfer during transient irradiation conditions remains a critical area of uncertainty in understanding nuclear reactor safety. This uncertainty impacts analytical predictions of both accident progression and fuel performance behavior. This uncertainty manifests itself in the use of extremely conservative models that widely bound the onset and duration of transition phenomena. Accurate and reliable descriptions of cladding-to-coolant heat transfer may also provide the primary building blocks to allow for a change in the current fuel safety criteria and enable safe operation at higher powers, which results in a substantial increase in the performance of current and future reactors. The complexity of the phenomena and the variety of thermal-hydraulic scenarios of interest requires establishing a mechanistic description of the behavior and a body of dedicated experimental data to validate the model.

This project developed an in-pile experiment for the TREAT facility to answer key questions regarding critical heat flux under prototypic irradiation with specific interest in transient conditions. The modeling scope of this project extended to the thermodynamic conditions in prototypic light water reactors. Experimental development and testing were focused on ambient coolant conditions. The research measured critical heat flux in the TREAT facility showing an approximate 2–4x increase from state-of-the-art predictions. To accomplish this goal, two unique-in-the-world capabilities were designed, constructed, and commissioned: (1) an in-pile experiment platform to study transient boiling in reactor; and (2) a high-power transient boiling experimental setup capable of running to relevant light water reactor conditions. Experiment design was guided by extensive modeling and simulation. The in-pile experiment platform was used in three separate assemblies for fourteen different experiments and demonstrated excellent performance. New machine learning analytical methodologies improved data extraction from the experiments (reduce uncertainties); these methods will benefit other experiments. A novel fission thermometer to improve understanding of energy deposited into the heater rod was developed, tested, and validated the model. The laboratory-based transient boiling setup was commissioned to show full functionality incorporating advanced diagnostics such as high-speed imaging and to experimentally challenging pressurized water reactor conditions.



(a)



(b)

(a) TREAT facility experiment design and modeling predictions. (b) Still frames from laboratory boiling tests; the right image shows a zoom on the surface revealing its local impacts.

TALENT PIPELINE:

- Alberto Cardenas Melgar, student at the University of Idaho
- Emory Brown, student at Oregon State University
- Jacob Gorton, student at the University of Tennessee, Knoxville
- Kevin Terrill, student at the University of Idaho
- Richard Hernandez, student at the University of Tennessee, Knoxville
- Seokbin Seo, postdoc at the University of Tennessee, Knoxville

AWARDS:

Best paper award to A. Melgar from the University of Idaho at the American Nuclear Society Student Conference 2021 for paper titled, “Design of the mechanical seal for the sight window used for an advance visualization instrument working in a high-pressure, high-temperature environment.”

2020 Innovations in Nuclear Technology R&D Award to R. Hernandez in 2021 for his paper that was published in Progress of Nuclear Energy in 2020 entitled, “Review of pool boiling critical heat flux (CHF) and heater rod design for CHF experiments in TREAT.”

NURETH-18 Young Professional Author Award to C. Jensen from INL in 2019 for his paper entitled, “Experimental design for transient in-pile boiling studies at the TREAT facility.”

PUBLICATIONS:

Hernandez, R., R. J. Armstrong, S. B. Seo, C. P. Folsom, C. B. Jensen, and N. R. Brown, "Sensitivity analysis of in-pile CHF experiments in the TREAT facility: Characterization of impacts of fuel system thermal properties," *Annals of Nuclear Energy* 165 (2022) 108645.

Seo, S. B., R. J. Armstrong, R. Hernandez, C. P. Folsom, C. B. Jensen, and N. R. Brown, "Sensitivity analysis of in-pile critical heat flux experiments in TREAT for characterization of reactivity-initiated accident power-transient effects," *Annals of Nuclear Energy* 161 (2021) 108448.

Hernandez, R., C. Folsom, N. Woolstenhulme, C. Jensen, J. Bess, J. Gorton, and N. Brown, "Review of pool boiling critical heat flux (CHF) and heater rod design for CHF experiments in TREAT," *Progress in Nuclear Energy* 123 (2020) 103303.

PRESENTATIONS:

Armstrong, R., et al., "Results of the CHF-SERTTA in-pile transient boiling experiments at TREAT," *Top Fuel 2021*, Oct. 24-28, 2021.

Melgar, A., et al., "Design of the mechanical seal for the sight window used for an advanced visualization instrument working in a high-pressure, high-temperature environment," *American Nuclear Society Student Conference*, April 8–10, 2021.

Armstrong, R., et al., "Calculation of critical heat flux using an inverse heat transfer method to support TREAT experiment analysis," *American Society of Mechanical Engineers International Conference on Nuclear Engineering (ICONE)-28-Power2020*, Aug. 4-5, 2020.

Hernandez, R., et al., "Incorporating axial boron gradients into a heater tube experiment for pool boiling CHF testing in TREAT," *American Society of Mechanical Engineers International Conference on Nuclear Engineering (ICONE)-28-Power2020*, Aug. 4-5, 2020.

Seo, S., et al., "Optimization study for uncertainty reduction of in-pile CHF experiments in TREAT," *American Society of Mechanical Engineers International Conference on Nuclear Engineering (ICONE)-28-Power2020*, Aug. 4-5, 2020.

Jensen, C., et al., "Preliminary results from in-pile transient boiling experiments at the TREAT facility," *American Nuclear Society Annual Meeting*, June 7-11, 2020.

Jensen, C., et al., "High speed boiling detector based on electrical impedance measurement for advanced in-pile boiling studies," *American Nuclear Society Winter Meeting*, Nov. 15-19, 2019.

Terrill, K., C. Jensen, C. Folsom, N. Woolstenhulme, D. Wachs, N. Smith, R. Christensen, R. Hernandez, and N. Brown, "Transient boiling during RIA: Out-of-pile testing and in-pile design for TREAT," *Top Fuel 2019*.

Hernandez, R., C. Folsom, N. Woolstenhulme, C. Jensen, and N. Brown, "Preliminary heater rod design for transient critical heat flux experiments in TREAT." *Transactions of 2019 American Nuclear Society Annual Meeting, American Nuclear Society*, June 9-13, 2019.

Jensen, C., C. Folsom, N. Woolstenhulme, N. Smith, K. Condie, K. Terrill, and R. Christensen, "Facility for Simulating Transient Boiling Behavior under PWR Conditions," *Transactions of 2019 American Nuclear Society Annual Meeting, American Nuclear Society*, June 9-13, 2019.

Hernandez, R., C. Folsom, N. Woolstenhulme, C. Jensen, J. Gorton, and N. Brown, "Sensitivity studies of heater rod design for transient critical heat flux experiments in TREAT," *18th International Topical Meeting on Nuclear Reactor Thermal Hydraulics, American Nuclear Society*, Aug. 18-23, 2019.

Terrill, K., R. Christensen, A. Fleming, and C. Jensen, "Advanced Instrumentation to Detect Boiling," *18th International Topical Meeting on Nuclear Reactor Thermal Hydraulics, American Nuclear Society*, Aug. 18-23, 2019.

Jensen, C., N. Woolstenhulme, C. Folsom, D. Kamerman, A. Fleming, and D. Wachs, "Experimental design for transient in-pile boiling studies at the TREAT Facility." 1*8th International Topical Meeting on Nuclear Reactor Thermal Hydraulics, American Nuclear Society*, Aug. 18-23, 2019.

Neutron Diffraction of Irradiated Nuclear Fuel

TOTAL APPROVED AMOUNT:
$1,298,400 over 3 years

PROJECT NUMBER:
18P37-007

PRINCIPAL INVESTIGATOR:
Chuting Tan

CO-INVESTIGATORS:
Aaron Craft, INL
Scott Moore, INL
Bryan Chakoumakos, Oak Ridge National Laboratory
Matthias Frontzek, Oak Ridge National Laboratory

COLLABORATORS:
Massachusetts Institute of Technology
Oak Ridge National Laboratory
Technical University of Munich
Los Alamos Neutron Science Center

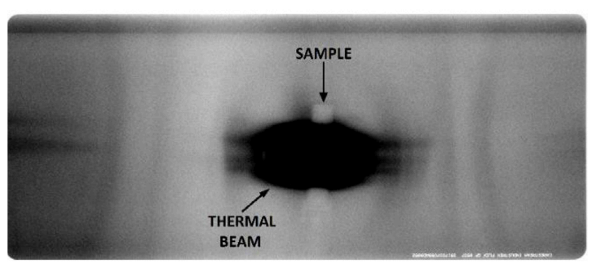
First-of-a-kind system enables characterization of highly radioactive materials that benefits nuclear fuel and materials development programs.

Despite the great interest in using neutron scattering techniques in nuclear materials and fuels research, the large-scale user facilities at laboratories worldwide currently do not have the programs, staff, or infrastructure to allow neutron scattering techniques to be applied to highly radioactive samples. Only recently has neutron diffraction started being explored as a tool for neutron irradiated materials. Even a rudimentary neutron diffraction capability for highly radioactive nuclear materials would be unique in the world and would introduce significant new access to materials science measurements for the discovery and diagnosis of nuclear material behavior.

A unique aspect of INL is its ability to routinely handle irradiated nuclear fuels and materials. A facility that: (1) specializes in handling highly irradiated materials; (2) has the capability to ship and receive highly irradiated materials globally; (3) has a reactor capable of generating a neutron beam with energies suitable for neutron scattering techniques; and (4) has an accessible neutron beam in which radioactive samples may be introduced, will enable first-of-a-kind research and international leadership in this area. The neutron beams at INL's Neutron Radiography Reactor meet these requirements and could enable neutron diffraction of irradiated nuclear fuels and materials.

This research demonstrated a first-of-a-kind proof-of-principle neutron diffraction system for examining highly radioactive materials at INL's Neutron Radiography Reactor. The team is pursuing funding to implement the capability at the Materials and Fuels Complex at INL. This capability will benefit nuclear fuel and materials development programs and provide data to benchmark and validate modeling and simulation codes that will accelerate the qualification process by requiring fewer experimental measurements.

(a)



(b)

(a) Feasibility study instrumentation in the Neutron Radiography Reactor's North Radiography Station. (b) Neutron radiograph of a nickel powder standard showing the sample, the incident neutron beam, and the diffracted beam.

TALENT PIPELINE:

- Chuting Tan, INL postdoc
- William Chuirazzi, INL
  Russell L. Heath Distinguished Postdoc

AWARD:

Best poster award: "Post-irradiation examination with neutrons at Idaho National Laboratory," *Neutron Science in Support of Nuclear Power* (2019).

PUBLICATION:

Vogel, S. C., M. A. M. Bourke, A. E. Craft, J. M. Harp, C. T. Kelsey, J. Lin, A. M. Long, A. S. Losko, P. Hosemann, K. J. McClellan, M. Roth, and A. S. Tremsin, "Advanced postirradiation characterization of nuclear fuels using pulsed neutrons," *JOM 72*(1) (2020) 187-196.

PRESENTATIONS:

Tan, C., C. Sun, X. Zhang, J. R. Bunn, E. A. Payzant, Y. Zhang, and H. Z. Bilheux, "Characterizing the effects of varying additive manufacturing parameters on stainless steel 316L," *American Conference on Neutron Scattering* (2020).

Tan, C., "Nuclear material diffraction instrument concept—Science," *Second Target Station Workshop*, Oak Ridge National Laboratory (2020).

Moore, S., "Nuclear material diffraction instrument concept—Instrumentation," *Second Target Station Workshop*, Oak Ridge National Laboratory (2020).

Tan, C., and W. Chuirazzi, "Post-irradiation examination with neutrons at Idaho National Laboratory," *Neutron Science in Support of Nuclear Power* (2019).

Development of New Experimental Capability based on Focused Ion Beam Micromachining and Investigation of Thermophysical Properties of Nuclear Materials at Micro and Mesoscale

TOTAL APPROVED AMOUNT:
$741,250 over 3 years

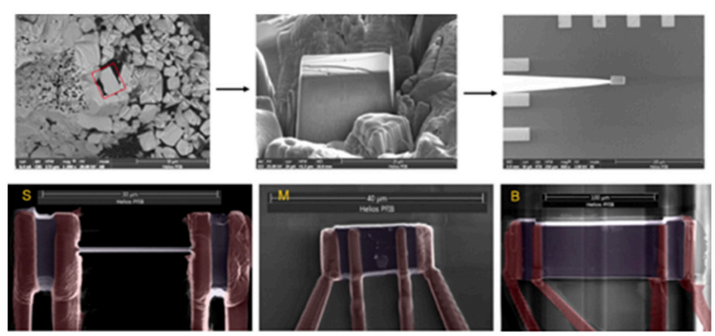
PROJECT NUMBER:
18P37-008

PRINCIPAL INVESTIGATOR:
Krzysztof Gofryk

CO-INVESTIGATORS:
Daniel Murray, INL
Luca Capriotti, INL

INL scientists elucidate fundamental properties of actinide materials and fuels at a microscale that can simplify material synthesis and advance transport characterization in future studies.

Advanced next-generation reactors demand a solid fundamental understanding of the physical properties of actinide materials and fuels, including transuranic elements. Despite intensive theoretical and experimental efforts, the effect of 4*f*- and especially 5*f*-electrons (electrons in the 5*f* orbital characteristic of actinide materials, such as uranium, neptunium, plutonium, americium, etc.) transport properties, and their interplay with micro- and meso-scale structures, such as grain boundaries, defects, and/or fission products, are still not well understood. The focused ion beam (FIB) instrument enables detailed microstructural characterization (phase identification, grain size/orientation, chemistry, etc.), extraction of single crystal samples, micromanipulation of specimens and other materials, and deposition of conductive materials. The FIB technique has been applied to measure electrical properties, but its use to directly probe thermal conductivity and other transport properties over a wide temperature and magnetic field ranges is novel with no such capability currently existing worldwide. The FIB, in conjunction with the newly developed three omega technique, will allow direct measurement of thermal transport properties of nuclear materials at the micro- and meso-scale. Researchers used this new experimental setup to study the thermophysical behaviors of various metallic and oxide nuclear materials, pure minor actinides, and the impact of microstructural features on these properties. This new experimental capability will be essential for the future validation of existing theoretical models and simulations, ranging from the nano- and micro-scale ab initio (density functional theory, molecular dynamics) to the meso-scale (Multiphysics Object-Oriented Simulation Environment (MOOSE)/MARMOT code).



(top) Electron image of a polycrystalline sample containing SmB_6 and SmB_4. The cubic SmB_6 is of main interest (SmB_6 grain is marked by the red rectangle) due to the presence of topological insulating states. (bottom) Images of plasma FIB prepared SmB_6 single crystals. The dimension of the small (S), midsize (M), and big (B) samples are 20x0.6x7 mm^3, 8x7x5 mm^3, and 110x70x22 mm^3, respectively. Each sample has four platinum contacts deposited by the plasma FIB method. The outer two contacts are current leads, and the inner two contacts are voltage leads.

TALENT PIPELINE:

- Narayan Poudel, INL postdoc
- Firoza Kabir, student at the University of Central Florida

PUBLICATIONS:

Poudel, N., D. Murray, J. Jeffries, and K. Gofryk, "Size dependent transport properties in micron-size crystals of topological Kondo insulator SmB_6" in preparation, intended for *Nature's Scientific Reports* (2021).

Poudel, N., J. Jeffries, and K. Gofryk, "Boundary scattering in micro-size crystal of topological Kondo insulator SmB_6" under review with *Applied Physics Letters*.

Hosen, M. M., G. Dhakal, B. Wong, N. Poudel, B. Singh, K. Dimitri, F. Kabir, C. Sims, S. Regmi, W. Neff, D. Murray, F. Weickert, K. Gofryk, O. Pavlosiuk, P. Wisniewski, D. Kaczorowski, A. Bansil, and M. Neupane, "Observation of gapped state in rare-earth monopnictide HoSb," *Nature's Scientific Reports* 10 (2020) 12961.

Kumar, N., C. Muniraju, R. Baral, Y. Tian, R. Li, N. Poudel, K. Gofryk, N. Barišić, B. Kiefer, J. H. Ross Jr., and H. S. Nair, "Magnetocaloric effect in a frustrated gd-garnet with no long-range magnetic order," *Inorganic Chemistry* 59(20) (2020) 15144-15153.

Episcopo, N., P.-H. Chang, T. W. Heitmann, K. Wangmo, J. M. Guthrie, M. Fitta, R. A. Klein, N. Poudel, K. Gofryk, R. R. Zope, C. M. Brown, and H. S. Nair, "Magnetic structure, excitations, and short-range order in honeycomb $Na_2Ni_2TeO_6$," *Journal of Physics: Condensed Matter* 33(37) (2021) 375803.

Dahal, R., L. Z. Deng, N. Poudel, M. Gooch, Z. Wu, H. C. Wu, H. D. Yang, C. K. Chang, and C. W. Chu, "Tunable structural phase transition and superconductivity in the Weyl semimetal $Mo_{1-x}W_xTe_2$" *Physical Review B* 101 (2020) 140505(R).

Baral, R., H. S. Fierro, C. Rueda, B. Sahu, A. M. Strydom, N. Poudel, K. Gofryk, F. S. Manciu, C. Ritter, T. W. Heitmann, B. P. Belbase, S. Bati, M. P. Ghimire, and H. S. Nair, "Signatures of low-dimensional magnetism and short-range magnetic order in Co-based trirutiles" Physical Review B 100 (2019) 184407.

PRESENTATIONS:

Poudel, N., M. Jaime, F. Weickert, J. Jeffries, and K. Gofryk, "Unusual magnetic anisotropy in uranium monosulfide single crystals," *74th Calorimetry Conference*, Santa Fe, NM, USA, July 28–Aug. 1, 2019.

Poudel, N., and K. Gofryk, "Magnetic anisotropy in uranium monosulfide single crystals probed by the torque magnetometry," INL postdoc appreciation week poster display (2019)

Poudel, N., D. Murray, J. Jeffries, and K. Gofryk, "Electrical and thermal transport properties of micron-size crystals of topological Kondo insulator, samarium hexaboride (SmB_6)," *Materials Research Society Spring Meeting 2019*, Phoenix, AZ, USA, Apr. 22–26, 2019.

Poudel, N., M. Jaime, F. Weickert, J. Jeffries, and K. Gofryk, "Magnetostriction and magnetic torque measurements of uranium monosulfide single crystals," *American Physical Society (APS) March 2019 Meeting*, Boston, MA, USA, Mar. 4–8, 2019.

Poudel, N, and K. Gofryk, "Transport measurements using AC techniques," INL/Bristol Bilateral Workshop, Idaho Falls, ID, USA (2018).

Accelerated Nuclear Materials and Fuel Qualification by Adopting a First-to-Failure Approach

TOTAL APPROVED AMOUNT:
$1,540,442 over 3 years

PROJECT NUMBER:
19A39-012

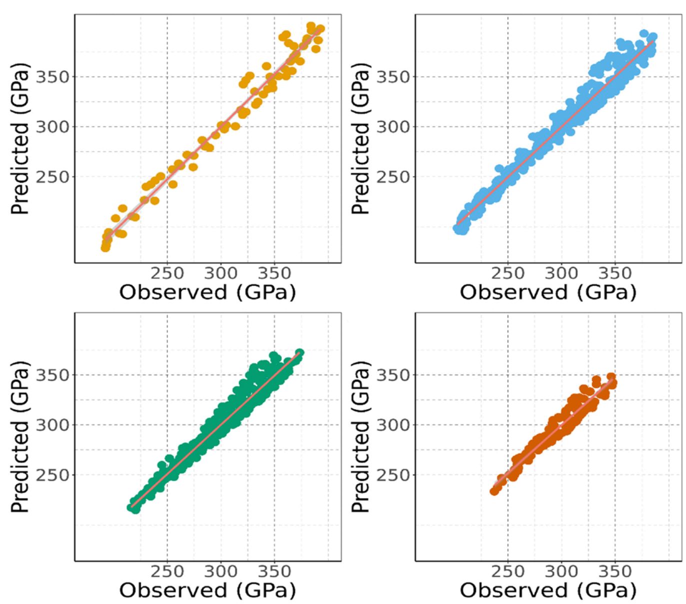
PRINCIPAL INVESTIGATOR:
Seongtae Kwon

CO-INVESTIGATORS:
David Swank, INL
Jeffery Aguiar, INL
Yongfeng Zhang, INL
Pratik Dholabhai, Rochester Institute of Technology
Taylor Sparks, University of Utah
Tolga Tasdizen, University of Utah

COLLABORATORS:
CalNano, Inc.
Sandia National Laboratory
University of Michigan

Combinatorial-based predictions using machine learning can shorten the nuclear materials selection cycle by half.

Researchers developed and demonstrated combinatorial workflows for materials alloy design through combined experimental and modeling research. Modeling, high-throughput fabrication, and predictive data analytics shortened the development cycle for structural nuclear materials selection. The current development process for nuclear materials involves a series of time-intensive steps and testing cycles. These methods suffer from ineffective early down-selection, requiring additional testing and qualification. The combination of several established and emerging methodologies promises significant opportunities for innovation that can shorten the cycle by as much as 50%. Using modeling and large-data analysis tools, researchers adopted a combinatorial methodology as a proof-of-principle development of a new class of alloys. Employing this approach hinged on the ability to organize, classify, and screen candidate materials and available data utilizing multi-scale modeling and large-data analysis, including deep, recursive, and transfer learning approaches. The approach began with combinatorial multi-scale modeling and scaled into extensive data analysis to identify candidate compositions for down-selection. The latter underwent a first-to-failure testing regime based on defined attributes critical for success from an early stage. Subsequent enhancements and comprehensive testing focused on a limited number of alloys with less risk in less time, demonstrating a new approach for material qualification. Each component of the method is individually novel. Combined, they promise significant opportunities for continued innovation and lasting impact in the broader materials community.



Machine learning offers excellent prediction accuracy. These materials properties have one non-linear term (cohesive energy) and three linear features (radius, valence electron concentration, shear modulus).

TALENT PIPELINE:

- Danielle Beatty, student at University of Utah
- Hayden Johnson, student at University of Utah
- Krithika Iyer, student at University of Utah
- Marcus Parry, student at University of Utah
- Michael Grant, student at Rochester Institute of Technology

PRESENTATIONS:

Parry, M., D. Beatty, K. Iyer, T. Tasdizen, T. Sparks, and J. Aguiar, "Combinatorial alloy fabrication and synergies with predictive frameworks for high-entropy alloy design and down-selection," *5th World Congress on Integrated Computational Materials Engineering (ICME) 2019*, Indianapolis, IN, USA, July 21–25, 2019.

Dholabhai, P., M. Grant, K. Iyer, T. Tasdizen, Y. Zhang, and J. Aguiar, "Developing a combinatorial modeling approach to multi-scale modeling and predictions for high entropy alloy design," *5th World Congress on Integrated Computational Materials Engineering (ICME) 2019*, Indianapolis, IN, USA, July 21–25, 2019.

Beatty, D., M. Parry, S. Kwon, K. Iyer, T. Tasdizen, T. Sparks, and J. Aguiar, "High-entropy alloy design, exploration, and down-selection using combinatorial fabrication and characterization for nuclear applications," *1st World Congress on High Entropy Alloys (HEA) 2019*, Seattle, WA, USA, Nov. 17–20, 2019.

Dholabhai, P., M. Grant, K. Iyer, T. Tasdizen, Y. Zhang, and J. Aguiar, "Multi-scale framework for predicting mechanical properties in high entropy alloys," *1st World Congress on High Entropy Alloys (HEA) 2019*, Seattle, WA, USA, Nov. 17–20, 2019.

Developing Multi-Scale, Rapid, and Comprehensive Post-Irradiation Examination using the Focused Ion Beam Microscope

TOTAL APPROVED AMOUNT:
$821,500 over 3 years

PROJECT NUMBER:
19A39-024

PRINCIPAL INVESTIGATOR:
Daniel Murray

CO-INVESTIGATORS:
Mukesh Bachhav, INL
Wade Marcum,
Oregon State University

Integrating multiple characterization technologies accelerates materials development through faster data acquisition and correlation.

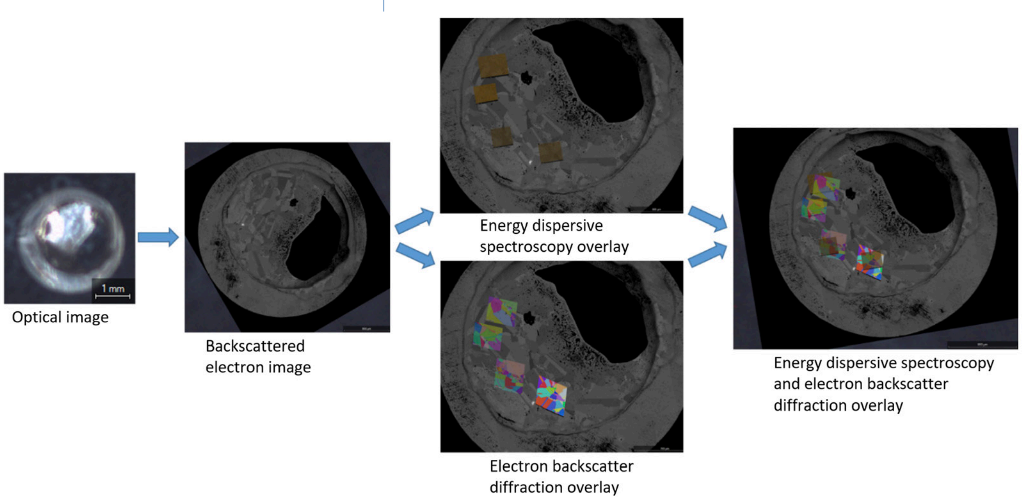
The current qualification cycle for nuclear fuels and materials is a multi-decade process that must be accelerated to bring many advanced reactor concepts into reality. This research focused on accelerating comprehensive characterizations of the microstructure and physical properties of irradiated fuels and materials providing higher quality correlated data. Current approaches to post-irradiation examination of materials are based on the use of single-purpose instruments, proceeding through a sequential process that provides incomplete characterization. Multiple samples are laboriously prepared and moved from instrument to instrument in an inefficient and uncorrelated process flow. Modern instrumentation with the capability for multi-modal analysis and automation offers the potential to radically improve the characterization data acquisition rate, the breadth of the data streams produced, and the data correlation. We developed technology based on dual-beam FIB and scanning electron microscopy platforms. These instruments incorporate separate ion and electron beams and include sample micromanipulation devices and tomographic imaging capabilities that provide the unique potential to incorporate many different characterization techniques into a single instrument. The resulting data streams are essential for understanding irradiation effects on materials and are ideal for the development and validation of mesoscale models, which are necessary for the development of accurate engineering-scale models.

TALENT PIPELINE:

- Robert Powers, student at Oregon State University

PRESENTATION:

Murray, D., F. Teng, N. Poudel, and K. Gofryk, "Advanced FIB/SEM characterization of nuclear materials in the Irradiated Materials Characterization Lab," Microscopy and Microanalysis 26(52) (2020) 1664–1665.



Correlated data acquisition workflow in the FIB platform.

Optical Fiber-based Fabry-Perot Interferometry for Fission Gas Detection

TOTAL APPROVED AMOUNT:
$182,000 over 3 years

PROJECT NUMBER:
19P43-007

PRINCIPAL INVESTIGATOR:
Austin Fleming

A new method measures fission gas released into the plenum region of nuclear fuel rods to improve thermal and mechanical modeling of reactors.

Understanding and predicting fission gas release is extremely important for both thermal and mechanical modeling of a reactor. Fission gases generally precipitate into bubbles that cause fuel swelling, resulting in pellet-cladding gap closure and mechanical interaction. Fission gas that releases to the fuel rod free volume (ultimately the plenum) causes pressure build-up and thermal conductivity degradation of the rod filling gas.

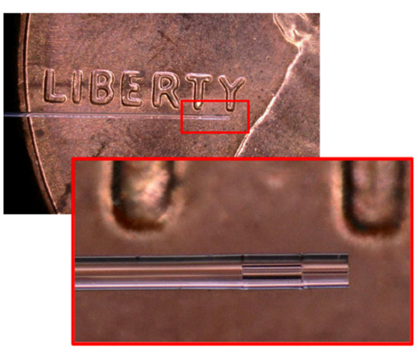
The objective of this research was to develop a sensor using optical fibers for in-pile detection of fission gas in the plenum region of fuel rods. The data from the new sensor can be used to validate the physics-based models of fission gas release and build-up in the plenum.

Currently, for pre-irradiated and re-fabricated test rodlets, the released fission gas (predominately xenon) is mixed with the backfilled helium. It is a technical challenge to measure the fission gas concentrations due to limited space and gas volume. Typical xenon measurements take advantage of the strong emission and absorption peaks in the near infrared wavelengths, which is impossible to implement inside the fuel cladding.

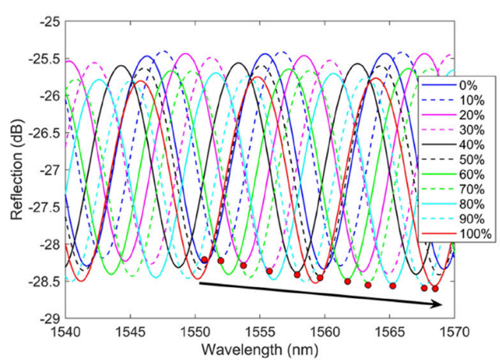
This research used xenon's high index of refraction as compared to helium to detect the concentration of fission gas in the plenum region, which is a novel application of this interferometric technique. The interferometry is based on a measurement of phase rather than amplitude, which minimizes the possible impact of radiation darkening on the measurement.

TALENT PIPELINE:

- Austin Fleming, INL
  Russell L. Heath Distinguished Postdoc
- Sohel Rana, student at Boise State University



(a)



(b)

(a) Fiber optic sensor made by splicing silica capillary tubes to single mode optical fiber. (b) Interference spectrum from sensor for varying argon and helium concentrations. The index of refraction can be determined from tracking an individual peak or the separation between peaks.

Seismic and Cost Assessment of Deeply Embedded or Buried Advanced Rectors with Seismic Isolation Strategies

TOTAL APPROVED AMOUNT:
$1,120,500 over 3 years

PROJECT NUMBER:
19A39-029

PRINCIPAL INVESTIGATOR:
Efe Kurt

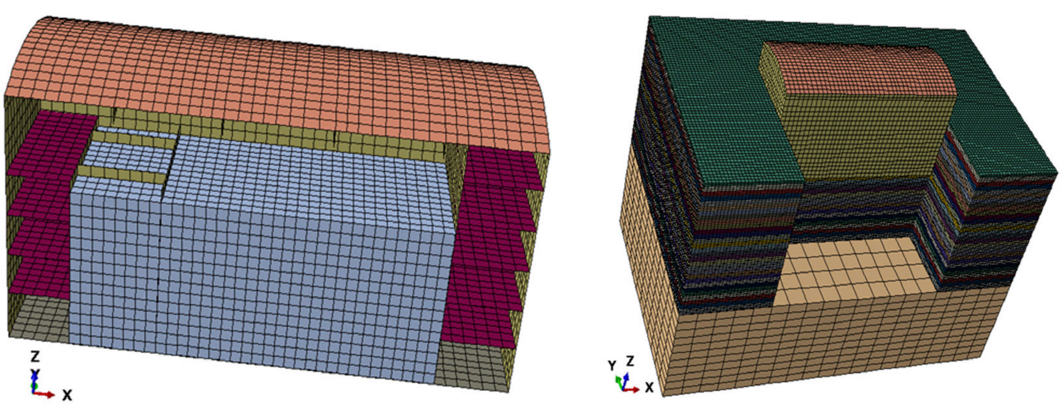
CO-INVESTIGATOR:
Justin Coleman, INL

COLLABORATOR:
Purdue University

Advancing understanding of the non-linear seismic behavior of deeply embedded advanced reactors to ensure their safe and cost-effective construction and operation.

Advanced nuclear power plants are being proposed with deeply embedded or buried reactor buildings. This approach stems from the technical and economic expectations that the overall safety of the plants will increase, while decreasing the cost by eliminating bulky shielding walls and structural dimensions. Along with deeply embedding the next-generation nuclear power plants, seismic isolators are considered for their unique safety and cost benefits to the systems, structures, and components. Currently, the performance of deeply embedded reactor buildings with or without seismic isolations is not well understood, especially under non-linear dynamic seismic environments.

This study investigated the seismic performance of deeply embedded advanced reactor concepts with different embedment depths of reactor buildings. The embedment ratios (embedment depth/reactor building height) were 0%, 25%, 50%, 75%, and 100%. Three-dimensional non-linear time-domain analyses were conducted for these embedment ratios with high nonlinearities in the soil while including the geometric nonlinearities occurring at the contact-interface region. The study focused on isolating safety-critical components using either lead-rubber or friction pendulum isolation. Small-scale physical experiments were conducted for the benchmark analysis of the interface behavior between the structures and the surrounding soil. Stochastic simulations were conducted to evaluate the most important soil material parameters in dynamic simulations.



(a) (b)

(a) Cut-away of the model with water explicitly modeled. (b) Cut-away of the model with the embedded structure.

TALENT PIPELINE:

- Samyog Shrestha, student at Purdue University

The observed general trend in the seismic response of the reactor, heat exchangers, and the buildings was that the acceleration amplitudes decrease with increased embedment depths. It was also shown that embedding the reactor buildings has positive effects on nuclear power plants economics, such as decreasing the thickness of the structures and reducing shutdowns during operations as compared to surface siting. A one-step non-linear numerical modeling approach was proposed and demonstrated for soil-structure and fluid-structure interactions of deeply embedded small modular reactors. Sloshing of models was validated with the available experiment data from the literature. One structural model was generated, and four seismic scenarios were considered based on embedment and how the water is modeled. When considering the response in the embedded and surface-founded structure that has the water mass distributed on the pool walls and the base, both models have a peak response at a frequency shifted down from a significant structural modal response. This is reasonable given that the full model includes water mass added to this location in the pool wall. When considering the response in the embedded and surface-founded structure that has the water explicitly meshed, both models have a peak response near the soil natural frequency. These peaks are due to the inertial mass of the water interacting with the wall, thus showing the effects of explicit modeling of the fluid body.

PUBLICATIONS:

Kurt, E. G., and R. Spears, "Seismic performance of deeply embedded conceptual advanced reactors: Three-dimensional nonlinear soil-structure interaction analyses," *Nuclear Technology* 207(11) (2021) 1664–1686.

Shrestha, S., E. G. Kurt, K. Kim, A. Prakash, and A. Irfanoglu, "Effect of soil properties and input motion on-site amplification using validated nonlinear soil model," *Nuclear Technolog*y 207(11) (2021) 1639–1663.

Determining the Role of Fission Gas Bubble Pressure in Fuel Fragmentation During Off-normal Conditions: A Separate-effect Study

TOTAL APPROVED AMOUNT:
$973,500 over 3 years

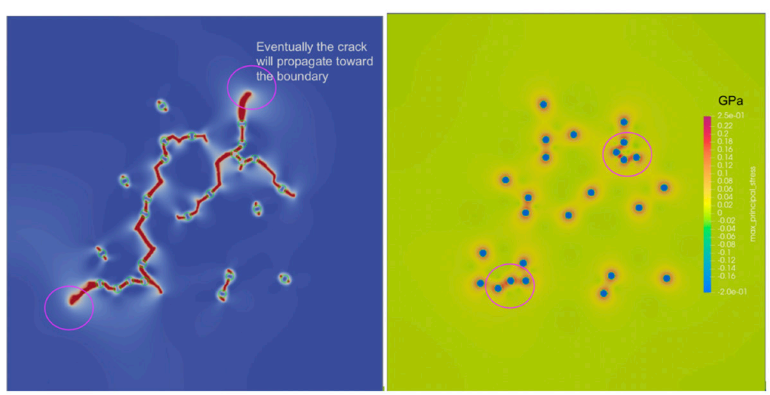
PROJECT NUMBER:
19A39-061

PRINCIPAL INVESTIGATOR:
Fabiola Cappia

CO-INVESTIGATORS:
Andrea Jokisaari, INL
Blair Grover, INL
Spencer Parker, INL
Jie Lian, Rensselaer Polytechnic Institute
Kun Mo, Argonne National Laboratory

By providing a separate-effect investigation to fragmentation issues, this project resulted in a remarkable understanding of phenomena causing structural modifications of ceramic fuels at high burnup.

A mechanistic understating of fuel behavior has been acknowledged as a primary need by the international scientific and industrial community in the past decade, particularly in advancing knowledge of the causality of input—output relationships for phenomena that impact the economics of the power plant and results in significant economic loss. One example is the limit in discharge burnup that is due to the concerns of fuel fine fragmentation during loss-of-coolant accident of high burnup fuel. In this project, we clarified the role of fission gas bubble pressure in fragmentation under an accelerated schedule. Instead of deploying integral irradiation tests that are expensive and take a long time to implement and execute, we deployed separate-effect studies under controlled conditions to address fuel safety issues and pair the experimental results with model developments. At the same time, sensitivity studies from simulations highlight the most critical parameters that drive fragmentation used to determine the experimental test matrix. Hence, we minimize the number of tests and ensure evaluation of the most significant conditions and increase the impact of testing results. From the experimental side, we induced microstructural modifications, such as recrystallization or bubble formation by engineering samples through advanced sintering techniques and ion-implantation, overcoming the issue of high radioactivity. The goal was achieved by partnering with Rensselaer Polytechnic Institute and Argonne National Laboratory and by developing a dedicated apparatus devoted to executing fast heating transients at INL, which expanded the laboratory's experimental capabilities. New models were implemented at engineering-scale and meso-scale levels to address the fuel fragmentation. The models were used to simulate the project testing conditions and fine-tune the test matrix.



(a) (b)

Phase-field fracture simulation of a random array of nanoscale bubbles in a 100 nm² range. (a) The propagated crack is shown in red. Notice how some bubbles display cracks that are not interconnected, while large interconnections exist between other bubbles. (b) The maximum principal stress in the material for bubbles with an overpressure of 200 MPa. The magnitude of the maximum principal stress between two bubbles depends on bubble spacing, with large values occurring between closely spaced bubbles.

TALENT PIPELINE:

- Mack Cullison, student at Oregon State University
- Mitchell Leibowitz, student at Boise State University
- Penghui Lei, student at Rensselaer Polytechnic Institute
- Tommaso Barani, student at Politecnico di Milano

PRESENTATIONS:

Cappia, F., G. Beausoleil, A. Winston, D. Murray, B. Miller, L. He, and F. Teng, "Investigation of high burnup ceramic fuel microstructure at Idaho National Laboratory," *The Minerals, Metals & Materials Society 2020 Annual Meeting & Exhibition*, San Diego, CA, USA, Feb. 23–27, 2020.

Cullison, M., F. Teng, D. Fraser, B. Kombaiah, K. Mo, J. Lian, T. Chen, and F. Cappia, "Microstructural analysis and micro-mechanical testing on Xenon-irradiated uranium dioxide," *The Minerals, Metals & Materials Society 2021 Annual Meeting & Exhibition*, Virtual, Mar. 15–18, 2021.

Moving Beyond Displacements Per Atom: A New Approach for Rapidly Quantifying Radiation Damage

TOTAL APPROVED AMOUNT:
$1,019,000 over 3 years

PROJECT NUMBER:
19A39-070

PRINCIPAL INVESTIGATOR:
Scott Middlemas

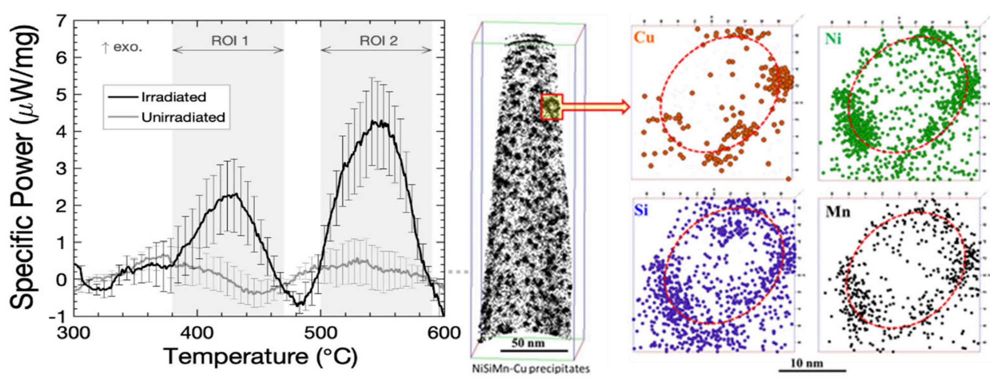
CO-INVESTIGATORS:
Daniel Murray, INL
Michael Short, Massachusetts Institute of Technology

New characterization technique offers a structural integrity metric for reactor pressure vessel steels, which are critical for evaluating nuclear reactor safety and lifetime.

The main objectives of this research were to develop and demonstrate a new experimental approach for directly measuring radiation damage of irradiated reactor materials that augments the calculation of displacements per atom and elucidates the recovery mechanisms during controlled annealing of these materials when coupled with advanced simulation and modeling. This was successfully accomplished by developing advanced scanning calorimetric techniques, advanced characterization of pre- and post-annealed samples, and advanced molecular dynamics and kinetic Monte Carlo models of the irradiated materials studied.

This research resolved and interpreted previously unobservable defects in irradiated metals through their energetic signatures. In neutron irradiated titanium, we investigated the final radiation damage recovery stage using differential scanning calorimetry (DSC) and reported first-of-a-kind measurements of stored energy from radiation damage in titanium. Additionally, we discovered two distinct exothermic release peaks indicating the recovery mechanism involves two sub-stages, whereas the previous radiation damage recovery model predicted one. Molecular dynamics simulations of defect annealing explored the corresponding defects for each process and were directly correlated to the experimental DSC results.

In a neutron-irradiated reactor pressure vessel, DSC and Flash DSC annealing experiments were performed and correlated with the pre- and post-annealed primary elemental distribution and defect structure using atom probe tomography, transmission electron microscopy, and positron annihilation spectroscopy. DSC experiments were performed on boron-doped 316 steel and ion-irradiated silicon carbide cladding materials.



(a) (b)

(a) DSC measures defect energy release upon annealing. (b) Atom probe tomography reveals key microstructural information of neutron irradiated reactor pressure vessel steel.

TALENT PIPELINE:

- Charles Hirst, student at Massachusetts Institute of Technology
- Kangpyo So, postdoc at Massachusetts Institute of Technology
- Penghui Cao, postdoc at Massachusetts Institute of Technology

We believe that precision DSC experiments and correlative atomistic simulations should be deployed along with other current powerful characterization techniques to yield insight into the structure-property relationships within materials. Today, this concept is in its infancy, but with further development, this strategy has significant potential for linking structure and properties in many areas of materials science.

PRESENTATIONS:

Hirst, C. A., R. S. Kemp, J. Li, M. Short, and S. Middlemas. "Revealing hidden defects via stored energy measurements of radiation damage," *The Minerals, Metals & Materials Society 2022 Annual Meeting & Exhibition*, Anaheim, CA, USA, Feb. 27–Mar. 3, 2022.

Hirst, C. A., P. Cao, and M. Short, "Direct measurement of radiation damage through the energy stored in defects: Simulations and experiments," *The Minerals, Metals & Materials Society 2020 Annual Meeting & Exhibition*, San Diego, CA, USA, Feb. 23–27, 2020.

Hirst, C. A., P. Cao, J. Li, R. S. Kemp, and M. Short, "Developing a method to measure radiation damage in metals using calorimetry," *6th Flash DSC Conference*, Zurich, Switzerland, Nov. 25–27, 2019.

Hirst, C. A., P. Cao, R. S. Kemp, and M. Short, "Developing a method to quantify radiation damage using stored energy: Simulations and experiments," *Materials in Nuclear Energy Systems (MiNES)*, Baltimore, MD, USA, Oct. 6–10, 2019.

Hirst, C. A., M. Short, P. Cao, R. Connick, and R. S. Kemp, "Developing a method to quantify radiation damage by measuring stored energy," *5th Nuclear Materials Conference,* Seattle, WA, USA, Oct. 14–18, 2018.

Archiving Experimental Breeder Reactor-II Metallic Fuel Test Data Using the Nuclear Energy Advanced Modeling and Simulation Program to Accelerate Fast Reactor Fuel Qualification

TOTAL APPROVED AMOUNT:
$1,137,000 over 3 years

PROJECT NUMBER:
19A39-103

PRINCIPAL INVESTIGATOR:
Douglas Porter

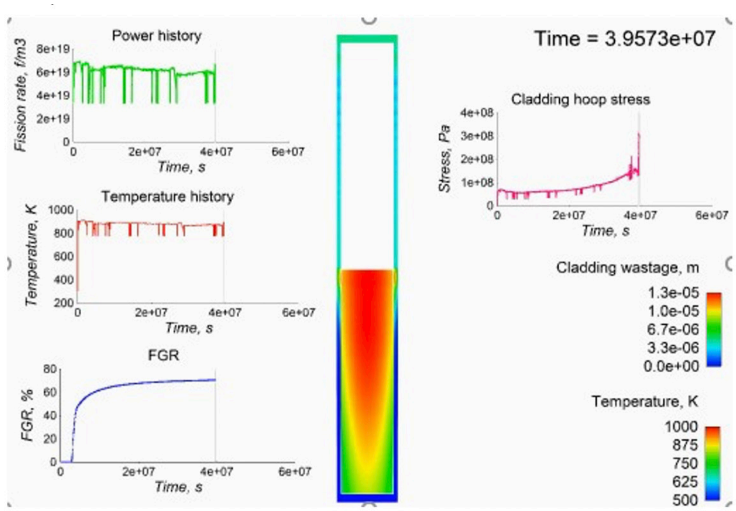
CO-INVESTIGATORS:
Andrei Gribok, INL
Nancy Lybeck, INL
Pavel Medvedev, INL

COLLABORATORS:
Argonne National Laboratory
Pacific Northwest National Laboratory

Linking multi-physics models with a centralized database containing historical experimental information accelerates nuclear fuel development and qualification.

Qualification of a fuel type for use in a new nuclear design can be very complicated and time-consuming, sometimes taking several decades. Combining data from experiments performed on similar fuel designs for different reactors into a single database allows those experimental results to be used to help qualification of the new fuel design. The data can also be used to validate the accuracy of fuel performance models. The database and model would be linked together so that design parameters for the new reactor fuel can be used to search the database for appropriate data for validation.

The goal of this research was to select an existing database, improve it as needed, and demonstrate the database and model linkage for a new fuel design. The sodium-cooled fast reactor was chosen as the example and the associated metal fuel database further selected because much of it had already been collected under the earlier Integral Fast Reactor Materials Information System program. New fuel performance modeling used by the Nuclear Energy Advanced Modeling and Simulation program was also selected for the demonstration. Argonne National Laboratory had already compiled the information into a new database, the Fuels Irradiation & Physics Database. A collaborative effort between the two laboratories resulted in successful linkage between the model and the database. It was shown that these data could be analyzed statistically to provide measures of accuracy of the model, which is required for use in fuel qualification and safety analysis support. Machine learning and image analysis techniques were applied to some of the rough data to enhance its usefulness. For example, radiography of irradiated test fuel pins was examined to extract specific information from the radiographs, such as fuel swelling.



A representation of some of the performance data that can be tracked and calculated for a single fuel pin from the database.

TALENT PIPELINE:

- Travis Wright, student at Texas Tech University
- Abhilasha Lokannavar, student at North Carolina State University
- Patrick Eblen, student at Idaho State University
- Ammon Black, student at Utah State University
- Kyle Paaren, INL postdoc

The original database contained post-irradiation data obtained from nearly 2,000 test fuel pins from the Experimental Breeder Reactor-II (EBR-II) and covered a wide variety of fuel and cladding compositions, operating temperatures, and lifetimes. Using the new database template, researchers also gathered information for experiments performed in the Fast Flux Test Facility as part of qualification efforts to include metal fuel driver in the Fast Flux Test Facility. The Fast Flux Test Reactor experiment used fuel pins with a core height nearly three times that of EBR-II, a height that more closely matches that of recently proposed reactors. With the help of the documentation of analyses performed at Pacific Northwest National Laboratory for the physics and other operational information and the post-irradiation examination collected at INL, the required information was put into a structured query language database that could later be added to the Fuels Irradiation & Physics Database. BISON modeling analysis demonstrated the success of these efforts, which are relevant to many new startup reactor types. For example, DOE and Arc Clean Energy have expressed interest for the Versatile Test Reactor and the ARC-100 advanced small modular reactor, respectively.

PUBLICATIONS:

Paaren, K. M., M. Gale, M. J. Kerr, P. G. Medvedev, and D. L. Porter, "Initial demonstration of automated fuel performance modeling with 1,977 EBR-II metallic fuel pins using BISON code with FIPD and IMIS databases," *Nuclear Engineering and Design* 382 (2021) 111393.

Paaren, K. M., N. Lybeck, K. Mo, P.G. Medvedev, and D. L. Porter, "Cladding profilometry analysis of EBR-II metallic fuel pins with HT9, D9, and SS316 cladding," *Energies* 14(2) (2021) 515.

Gribok, A. V., D. L. Porter, K. M. Paaren, M. D. Gale, S. C. Middlemas, and N. J. Lybeck, "Automatic information extraction from neutron radiography imaging to estimate axial fuel expansion in EBR-II," *Journal of Nuclear Materials* 557 (2021) 153250.

PRESENTATION:

Wright, T. R., and D. L. Porter, "Utilization of several post-irradiation measurement techniques to determine axial growth in EBR-II fuel," *American Nuclear Society Winter Meeting 2019*, Washington D.C., USA, Nov. 17–21, 2019.

Light Element Analysis of Nuclear Fuels by Electron Probe Microanalysis

TOTAL APPROVED AMOUNT:
$128,000 over 3 years

PROJECT NUMBER:
19A41-025

PRINCIPAL INVESTIGATOR:
Karen Wright

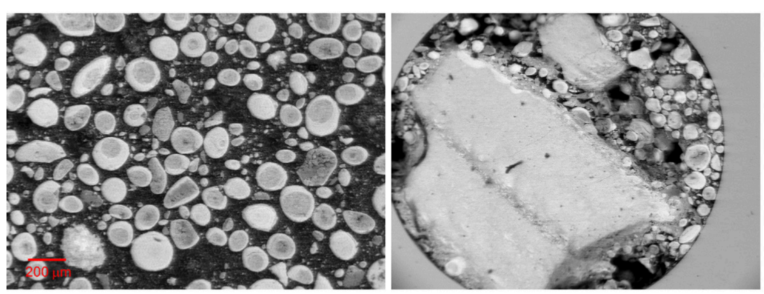
CO-INVESTIGATORS:
Fidelma Di Lemma, INL
Jacob McMurray, Oak Ridge National Laboratory

COLLABORATOR:
University of Barcelona

A new capability furthers understanding of how oxide fuel performs in a reactor and how fission products migrate within the fuel.

The purpose of this project was to develop a cost-effective method to use electron probe microanalysis (an X-ray based technique) to quantify the carbon and oxygen quantities with the spatial resolution of a few micrometers in uranium-based nuclear fuel. In oxide fuels, the oxygen-to-uranium ratio evolves during irradiation and affects fuel properties such as thermal conductivity and fission product oxidation state. Decreasing thermal conductivity results in hotter fuel, which enhances fission product transport and increases fuel swelling. Increased fuel swelling can lead to cladding compromise or failure. Therefore, it is important to understand how the oxygen-to-uranium ratio varies spatially across the sample and how it varies with increased temperature and burnup. The average oxygen-to-uranium ratio for an irradiated fuel pin can be measured using bulk analysis techniques. Being able to quantify the oxygen-to-uranium ratio on the micrometer scale is novel.

Several objectives were achieved during this project, including determining the extent to which carbon and oxygen X-rays are absorbed by uranium, determining the accuracy that is possible when measuring oxygen in uranium fuel, and testing different X-ray matrix correction models, which are necessary to reduce the electron-physics-based artifact impact on data quality. After some preliminary experiments, high-quality uranium dioxide was obtained and used to determine oxygen X-ray absorption in uranium, to determine the mass absorption coefficient from that data, and to then test the newly acquired mass absorption coefficient on the measurement of non-stoichiometric uranium dioxide. In addition, issues challenging carbon quantification in uranium and steps to address these challenges were identified.



(a) (b)

(a) Uranium oxide (U_3O_8) grain mount calculated to have an oxygen k-alpha mass absorption coefficient in uranium of 7494. (b) U_3O_8 bulk sample calculated to have an oxygen k-alpha mass absorption coefficient in uranium of 6954.

TALENT PIPELINE:

- Karen Wright, student at Technical University Delft
- Mike Matthews, student at the University of Bristol

This research demonstrated that the electron probe microanalyzer can be used to distinguish small changes in oxygen-to-metal stoichiometry. Therefore, it is possible to use electron probe microanalysis to measure non-stoichiometric uranium dioxide and to use that data to determine diffusion coefficients for oxide species in oxide fuel. This should enable improved modeling capabilities for fission product transport in oxide fuels, thus improving predictions, particularly for off-normal events.

PRESENTATIONS:

Wright, K. E., "Determining mass absorption coefficients of O kα and C kα in uranium," *Quantitative Microanalysis Topical Conference*, Minneapolis, MN, USA, June 24–27, 2019.

Wright, K. E., "Electron probe microanalysis of actinide elements: Our past, present, and future," *Microscopy and Microanalysis 2019 Meeting*, Portland, OR, USA, Aug. 4–9, 2019 (Invited presentation).

Evaluating Thermal Properties of Advanced Materials

TOTAL APPROVED AMOUNT:
$304,680 over 3 years

PROJECT NUMBER:
19P43-009

PRINCIPAL INVESTIGATOR:
Tsvetoslav Pavlov

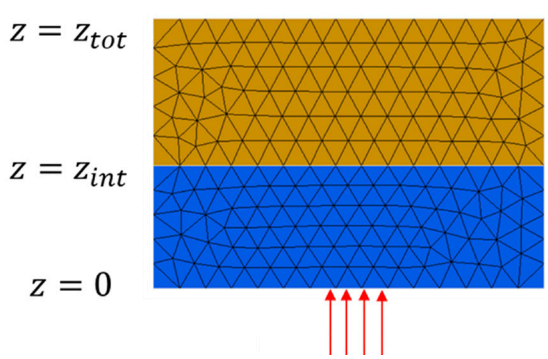
COLLABORATOR:
Rensselaer Polytechnic Institute

First-of-a-kind analysis tool enables accurate measurement of multiple thermophysical properties of specimens with asymmetric geometries.

Thermal conductivity, thermal contact conductance, specific heat, and total hemispherical emissivity are critical for predicting the fuel temperature during normal and off-normal reactor operation. Knowledge of these properties is imperative to licensing existing and novel fuel and cladding materials. Prior to this research, only thermal diffusivity was measured via the laser flash technique at INL, which measures the temperature increase on the rear side of a specimen induced by a laser pulse (flash) on the front side of the specimen. The conventional method uses sample thickness and the time instant at which the measured temperature is at half of its maximum value to determine thermal diffusivity.

This project enhanced this technique and its capabilities by developing a novel data analysis tool and applying it to new, advanced materials. This was accomplished by further developing a machine learning-based tool, as well as modifying the laser flash setup. The tool consists of a finite element model (FEM), a least-squares fitting algorithm and experimental data treatment algorithms. Model data is fit to experimental data by using specific thermophysical properties as optimization parameters. The new tool is based on the most recent studies in the field and will allow the determination of multiple thermophysical properties (e.g., thermal conductivity, specific heat, thermal diffusivity, thermal contact resistance, total emissivity) from a single laser flash measurement.

The project consisted of three main stages: (1) adapting the tool for measurement of asymmetric specimen—fuel fragments and curved cladding specimen; (2) modifying the laser flash facilities in a cost-efficient manner to harvest the full potential of the advanced tool—modifying laser flash components such sample holders and crucibles; and (3) applying the new methodology to various materials of interest. Furthermore, the tool is unique as it can be quickly modified for different specimen, such as encapsulation of liquid specimen for molten salt reactors.



Representation of the contact conductance FEM, which simulates the contact between tungsten (blue) and zirconium nitride (brown) via a contact conductance boundary condition at thickness $z = z_{int}$. The red arrows indicate the direction of the laser beam and heat flow.

TALENT PIPELINE:

- Tsvetoslav Pavlov, INL
  Russell L. Heath Distinguished Postdoc

AWARD:

Exceptional contributions award at INL's Materials and Fuels Complex

The main benefits of the new tool are: (1) a cost-effective approach – fast determination of multiple properties using existing facilities; (2) ability to design experiments in a fast and inexpensive manner using FEM; and (3) a state-of-the-art methodology that has the potential to make both scientific and commercial impact.

PRESENTATIONS:

Pavlov, T. R., "Towards improved measurement capabilities and better understanding of the thermo-physical properties of nuclear materials," *NuFuel Materials Modeling and Simulation for Nuclear Fuels Workshop 2019*, Zurich, Switzerland, Nov. 4–7, 2019.

Pavlov, T. R., "Thermophysical property characterization at INL," *Georgia Institute of Technology Seminar* (2019).

Pavlov, T. R., "Towards state-of-the-art thermophysical property characterization capabilities at INL," *Imperial College London Seminar* (2019).

Proton Recoil Effect on Light Output in Fast Neutron Scintillators

TOTAL APPROVED AMOUNT:
$120,000 over 1 year

PROJECT NUMBER:
20A1047-029

PRINCIPAL INVESTIGATOR:
William Chuirazzi

CO-INVESTIGATORS:
Aaron Craft, INL
Burkhard Schillinger, Technical University of Munich

Research to understand the physics of fast neutron scintillator screens lays the foundation for improved fast neutron imaging to help advance nuclear fuel development.

Fast neutron imaging has important implications for nuclear fuel development. Fast neutron imaging has thus far been limited to a poorer spatial resolution to ensure the scintillator screen outputs enough photons to render an image.

This project performed a parametric study to inform production of fast neutron scintillator screens that provide improved spatial resolution. The study consisted of custom fast neutron scintillators that isolate the effects of the converter and phosphor by using high-density polyethylene as a neutron converter and various phosphors to produce scintillation photons. The fast neutrons interact with the high-density polyethylene to create recoil protons, which then interact with the phosphor material to produce visible light.

Additionally, this project studied the underlying physical mechanisms that generate fast neutron scintillation to solve an apparent disagreement between common theory and widespread experimental measurements. Literature describes the range of a recoil proton in fast neutron scintillators to be ~50-300 µm. Recoil protons created from the interaction of the materials with INL's Neutron Radiography Reactor's beamline have an energy around 1-4 MeV. Therefore, the number of recoil protons should increase for high-density polyethylene thicknesses up to the range of the recoil protons (e.g., 50-300 µm) but level off after for thicker high-density polyethylene. In initial testing, however, a fast neutron scintillator with high-density polyethylene thickness varying from 0.25-3.0 mm demonstrated an increase in light output for thicknesses up to the maximum thickness of 3 mm, well beyond the 50-300 µm range of the proton recoil.

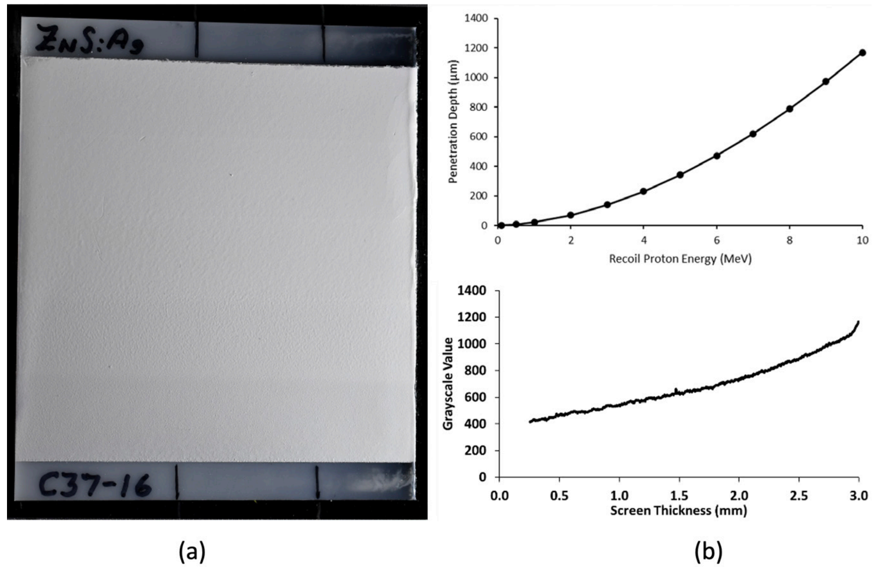
This research: (1) advances the study of the hypothesis that the current understanding of proton recoil's light output in fast neutron scintillators is incomplete and that proton build-up in high-density polyethylene causes it to act as an amplifier that produces a higher light output in fast neutron scintillators; (2) provides for experiments and analysis toward the early determination of the utility of optimized fast neutron scintillator composition; and (3) supports the conception and preliminary technical analysis of the experimental high-resolution fast neutron scintillator screens.

TALENT PIPELINE:

- William Chuirazzi, INL
  Russell L. Heath Distinguished Postdoc
- Jesus Mendoza, student
  at the Colorado School of Mines

INTELLECTUAL PROPERTY:

Chuirazzi, W., A. Craft, S. Cool, and B. Schillinger, "Fast neutron scintillator screens comprising layers, and related methods and systems," United States (U.S.) Patent Application No. 17/451,610 (Oct. 20, 2021).



(a) A fast neutron scintillator screen consisting of a wedged layer of high-density polyethylene coated with zinc sulfide an silver (ZnS:Ag) scintillator. (b, top) Simulation results showing the mean penetration depth of recoil protons in high-density polyethylene. (b, bottom) Experimental results of light output as a function of high-density polyethylene thickness.

Conception of an Enhanced Fast Neutron Test Capability in a Thermal Spectrum Reactor

TOTAL APPROVED AMOUNT:
$120,000 over 1 year

PROJECT NUMBER:
20A1049-002

PRINCIPAL INVESTIGATOR:
Nicolas Woolstenhulme

CO-INVESTIGATORS:
Bryon Curnutt, INL
Colby Jensen, INL
Kevan Weaver, INL

Three-dimensional nuclear transport modeling decreases the cost of sodium-cooled fast reactor fuel development by showing a pathway to simulate reactor conditions using existing fuel assemblies and the Advanced Test Reactor.

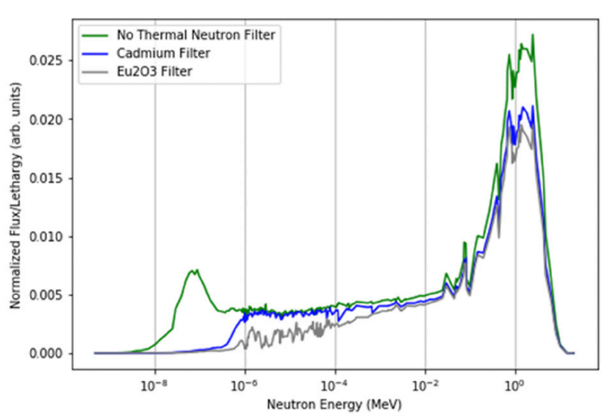
Development of advanced fuels and materials for fast spectrum nuclear reactors is an immense challenge with tremendous potential to maximize the value of advanced nuclear plants. Historical developments and modern materials show promise for overcoming these challenges, but the current lack of fast neutron test capabilities in the western world hampers meaningful progress. This project used modern modeling to investigate a new method for fast neutron irradiations in an existing thermal-spectrum reactor, the Advanced Test Reactor. This new method will build upon past ideas where fast flux is increased by surrounding the specimens with fissionable "booster fuel," but diverges from historical approaches by using an "off-the-shelf" fuel assembly as the booster fuel while leveraging advancements in three-dimensional simulation and geometries enabled by modern manufacturing. This concept has been termed the Advanced fast Reactor Concept in Thermal-spectrum Irradiation Capability (ARCTIC). These studies used modern modeling to investigate the hypothesis that ARCTIC would result in a worthwhile fast flux increase whose deployment in the Advanced Test Reactor would be timely in maturing fast spectrum research while the community awaits availability of a true fast spectrum test reactor.

TALENT PIPELINE:

- Keegan Pombier, student at University of Michigan

PUBLICATION:

Curnutt, B., N. Woolstenhulme, J. Nielsen, N. Oldham, K. Weaver, C. Jensen, and A. Fradeneck, "A neutronics investigation simulating fast reactor environments in the thermal spectrum advanced test reactor," *Nuclear Engineering and Design*, 387 (2022) 111623.



Normalized flux energy profile within ARCTIC with no thermal neutron filter, cadmium filter, and europium(III) oxide filter.

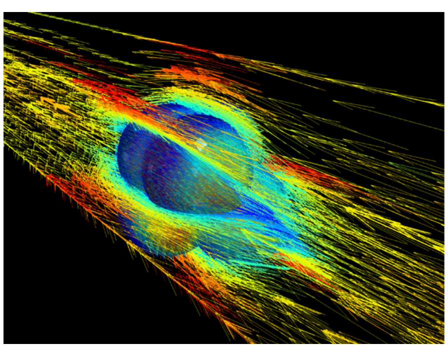
Multi-scale Thermal Properties Prediction in the Multiphysics Object-Oriented Simulation Environment via a General Boltzmann Solver

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PROJECT NUMBER:
20A1049-012

PRINCIPAL INVESTIGATOR:
Jackson Harter

CO-INVESTIGATORS:
Andrea Jokisaari, INL
Sebastian Schunert, INL
Shuxiang Zhou, INL



Heat flow around a cluster of xenon bubbles in a uranium dioxide lattice.

First principles modeling of thermal properties in solids allows researchers to narrow the field of possible materials and geometries for new thermoelectric devices, quantum computing components, and nanosensors, enabling substantial time and monetary savings.

The physics of thermal properties in solids are inherently multi-scale, arising from atomistic processes and interactions with the microstructure of a material, such as grain boundaries and point defects. Heat transport in solids via conduction occurs through the transport and scattering of electrons and phonons. The a priori forecasting of fundamental quantities, such as temperature distribution, heat flux, and thermal conductivity typically employs the Boltzmann transport equation to predict the macroscopic behavior of a materials system in terms of the microscopic dynamics of its heat carriers.

This project established a new MOOSE module, Boltzmann, dedicated to phonon and thermal electron transport. This is a deterministic transport method using the continuous FEM to resolve the geometric domain and discrete ordinates to resolve the angular domain. Thermal electron transport is coupled through a spatially averaged temperature in the Fermi-Dirac statistics, which describe the thermodynamic equilibrium distribution of electrons. The transport algorithm leverages these statistics as the Boltzmann transport equation to calculate the deviation from equilibrium, which leads to the description of a temperature field, heat flux, and electronic/thermal conductivity. This method was tested on two-dimensional films of silicon at room temperature based on the available material properties, but the method works the same for metallic materials. In addition, spectrally resolved phonon transport is implemented with the same spatial and angular discretization as thermal electron transport. A new algorithm and method also were developed that leverage the equilibrium Bose-Einstein statistics for non-interaction particles.

Boltzmann provides a platform to sustain increasingly sophisticated models of the collision physics inherent to the transport of the heat carriers. This software simulates thermal electron and phonon transport to predict fundamental thermal quantities based on ab initio material property data, which may be used at the engineering scale. This powerful capability provides a critical bridge across the length scales, allowing the prediction of material properties based on ab initio approaches. The simulation of coupled electron-phonon transport allows for a holistic depiction of the detailed, mechanistic process occurring at the atomistic- and nano-scales.

PUBLICATION:

Harter, J. R., T. S. Palmer, and P. A. Greaney, "Predicting mesoscale spectral thermal conductivity using advanced deterministic phonon transport techniques," Chapter 5 in *Advances in Heat Transfer*, Abraham, J. P., J. M Gorman, and W. J. Minkowycz (eds.), vol. 52 (2020).

Using Coupled Multiphysics Tools to Investigate Design-limiting Criteria for Molten Salt Reactors

TOTAL APPROVED AMOUNT:
$111,723 over 1 year

PROJECT NUMBER:
20A1049-015

PRINCIPAL INVESTIGATOR:
Sebastian Schunert

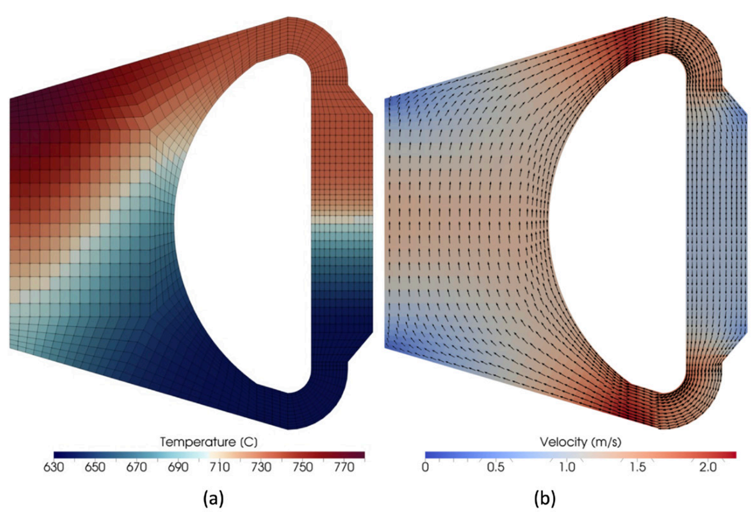
CO-INVESTIGATORS:
Abdalla Abou-Jaoude, INL
Nicolas Martin, INL
Paolo Balestra, INL

Multiphysics-based modeling and simulation of molten salt reactors enable efficient design efforts and improved safety.

The goals of this research were to: (1) perform first-of-a-kind, high-fidelity, transient modeling and simulation of molten salt reactors (MSRs); and (2) explore the impact of design choices on core behavior during an unprotected loss of forced cooling accident.

The first phase of the project involved the direct coupling of the Griffin and Pronghorn codes. Griffin is the MOOSE-based reactor multiphysics code created by joining INL (Rattlesnake and MAMMOTH) and Argonne National Laboratory (Multigroup Cross section generation Code cross-sections and PROTEUS neutronics) capabilities. Pronghorn is a multi-dimensional, coarse-mesh, thermal-hydraulics code for single-phase fluids. Both Griffin and Pronghorn are equipped to address the fundamental physics challenges posed by MSRs while keeping execution time below an hour on a desktop computer. Coupling the two codes for multiphysics analysis of MSR concepts was demonstrated as part of this project.

The second phase of the project involved testing the novel capabilities against an open-source benchmark for MSR. The results agreed well with other solutions provided by alternative codes. The third phase of the project then implemented these capabilities to a simplified MSR geometry and investigated the sensitivity of core design parameters on the steady-state behavior of an MSR. The last phase of the project dealt with design-limiting considerations for MSR, namely the impact of thermophysical property uncertainties on safety performance issues. Due to the lack of existing capabilities to irradiate and conduct post-irradiation examination uncertainties on salt-based fuel and industry interest in high burnup salts, assessing 'allowable' ranges in property uncertainties will be crucial to future MSR deployment. The project demonstrated a framework for evaluating the impact of thermophysical properties against multiphysics accident conditions in a MSR (e.g., unprotected loss of flow accident).



Steady-state temperature (a) and velocity (b) distributions in a molten salt fast reactor obtained from the coupled multiphysics Griffin/Pronghorn simulation with nominal material properties.

TALENT PIPELINE:

- Andrew Hermosillo, student at Texas A&M University
- Ramiro Freile, student at Texas A&M University

PUBLICATION:

Abou-Jaoude, A., S. Harper, G. Giudicelli, P. Balestra, S. Schunert, N. Martin, A. Lindsay, M. Tano, and R. Freile, "A workflow leveraging MOOSE transient multiphysics simulations to evaluate the impact of thermophysical property uncertainties on molten-salt reactors," *Annals of Nuclear Energy* 163, 2021, 108546.

PRESENTATIONS:

Balestra, P., A. Abou-Jaoude, N. Martin, S. Schunert, A. Hermosillo, and Y. Wang, "Multiphysics simulation of the Molten Salt Fast Reactor using Griffin and Pronghorn," *Mathematics & Computation* (2021).

Balestra, P., A. Abou-Jaoude, S. Schunert, A. Hermosillo, Y. Wang, and N. Martin, "Coupled multiphysics simulation of pool-type molten salt reactors using Griffin/Pronghorn," *American Nuclear Society Virtual Winter Meeting 2020*, Nov. 16-19, 2020.

Ion-Solid Interactions in Focused Ion Beam/ Secondary Ion Mass Spectrometry: Toward Nanoscale Trace Element and Isotopic Distribution Analysis in Irradiated Materials

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PROJECT NUMBER:
20A1052-018

PRINCIPAL INVESTIGATOR:
Daniel Murray

CO-INVESTIGATORS:
Xiaofei Pu, INL
Kurt Langworthy, University of Oregon
Stephen Golledge, University of Oregon

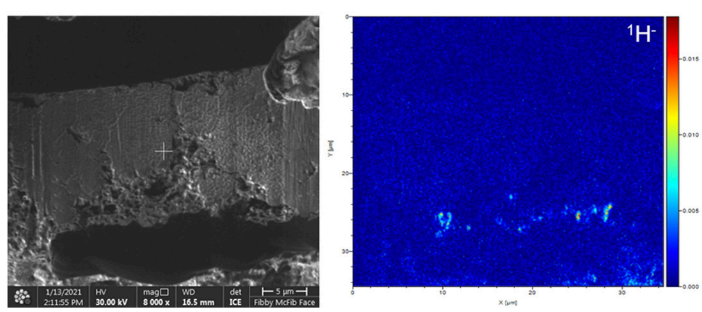
Proof-of-concept of a new tool opens possibilities for the development of materials for advanced battery applications.

Micro-analytical characterization provides critical insights to micro-, nano-, and atomic-scale processes that take place in advanced nuclear reactors. The scope of these insights is defined by the scope of instrumental capabilities that are available to researchers. This project conducted an in-depth study of ion-solid interactions in the new G4 Thermo Helios Hydra Plasma FIB (PFIB)/scanning electron microscopy/secondary ion mass spectroscopy (SIMS) system that is installed in the Irradiated Material Characterization Laboratory at INL. This instrument has an unprecedented combination of various analytical techniques and presents distinct opportunities for in situ analysis of irradiated materials, including all elements and their isotopes from hydrogen to californium at nanometer scale.

This study on ion-solid interaction in the new G4 PFIB-SIMS expanded fundamental understandings of this characterization technique and, thus, provided a larger scope of instrument capability in analyzing trace elements (<0.01 atomic percent in concentration), light elements (e.g., hydrogen, helium, lithium, beryllium, boron), and isotopic compositions in irradiated materials in situ with high spatial resolution. The G4 FIB-SIMS is unique in that it has four primary ion beams—nitrogen, oxygen, argon, and xenon—all available in a single instrument. In addition to investigations on the ion-solid interactions in a range of materials, we also demonstrated how the different primary ion beams complement each other in resolving signal interference and enable high-quality data analysis for a more inclusive range of isotopes. The work in this project was the pilot effort in filling knowledge gaps in multi-ion PFIB-SIMS, which is critical for using the full potential of this first of its kind, state-of-the-art instrument in innovative nuclear material research. This new analytical capability will also benefit other areas of materials science, such as research and development for advanced batteries.

PRESENTATION:

Murray, D., and X. Pu, "FIB-SIMS in a multi-ion source plasma FIB," *The Minerals, Metals & Materials Society Annual Meeting and Exhibition 2021*, Virtual, Mar. 15–18, 2021.



(a) (b)

Capability demonstration mapping hydrogen distribution in a lithium battery material. (a) Cross section of an interlayer phase. (b) The corresponding hydrogen map. This demonstrates the ability to map hydrogen distribution, which is important for many nuclear materials systems, and shows the extremely high resolution of the instrument. Features down to 100 nm and below can be seen in this map.

Heat Source Identification Using Inverse Optimization

TOTAL APPROVED AMOUNT:
$144,000 over 1 year

PROJECT NUMBER:
20A1052-022

PRINCIPAL INVESTIGATOR:
Lynn Munday

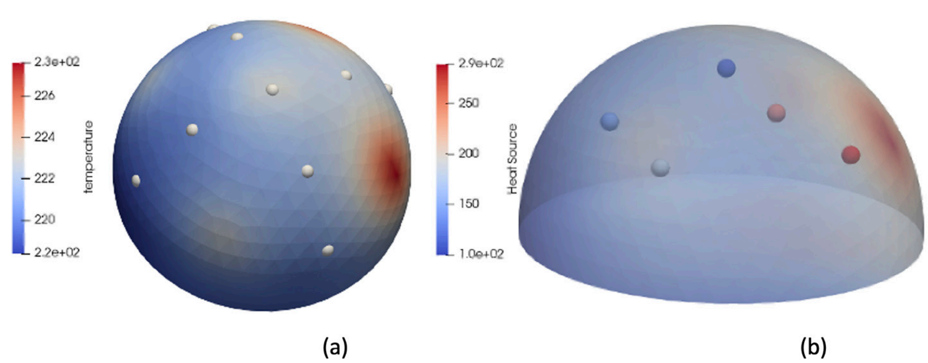
CO-INVESTIGATORS:
Chandrakanth Bolisetti, INL
Daniel Schwen, INL
Dewen Yushu, INL
Zachary Prince, INL
Murthy Guddati,
North Carolina State University

Multiphysics modeling and simulation determine heat source intensities when direct temperature measurements are not possible.

Modeling and simulation can accurately predict the temperature distribution in a structure from a known heat source. In this project, researchers examined the inverse problem—determining the properties of the heat source from a set of temperature measurements. The solution to these types of inverse parameter estimation problems can be written as a partial differential equation constrained optimization problem. The partial differential equation describes the coupled physics of the processes, and modeling and simulation can identify the parameters of the partial differential equation that most closely fit the measurements. This work implemented the optimization algorithms required for solving the inverse problem of reconstructing a heat source that produces a known temperature distribution using gradient-based optimization. This project developed a proof-of-concept design for a general gradient-based partial differential equation constrained optimization framework in our MOOSE-based application named InverSe OPtimizatiOn and Design (ISOPOD).

ISOPOD provides a MOOSE executioner interface to the Portable, Extensible Toolkit for Scientific Computation (PETSc) Toolkit for Advanced Optimization. The PETSc Toolkit for Advanced Optimization provides several Hessian, gradient, and gradient-free optimization algorithms that can be used in ISOPOD. ISOPOD provides gradient information to the Toolkit for Advanced Optimization by solving the adjoint of the original physics problem. The ISOPOD application implements this software in the context of existing MOOSE objects, including a new executioner, reporter, Dirac kernel, and MultiApp with transfers. Documentation and testing of ISOPOD is done within the MOOSE markdown and testing system with plans of merging this into the main MOOSE code base, providing users with an entirely new type of modeling and simulation capability. The multiphyisics nature of MOOSE will make this new capability extensible to parameterization of other multiphysics processes. For example, this work can be extended to other optimization problems, such as topology optimization and material design.

INTELLECTUAL PROPERTY:
ISOPOD is open-source software in GitHub.



Steady-state temperature (a) and velocity (b) distributions in a molten salt fast reactor obtained from the coupled multiphysics Griffin/Pronghorn simulation with nominal material properties.

Development of an Innovative Testing Framework to Assess Crack Initiation Assisted by Corrosion-Fatigue

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PROJECT NUMBER:
20A1052-025

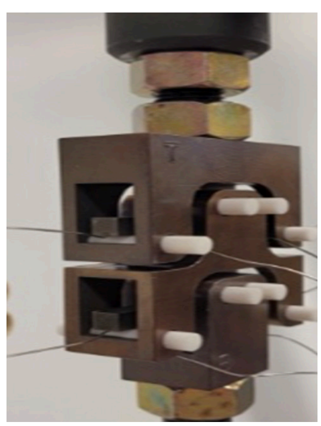
PRINCIPAL INVESTIGATOR:
Boopathy Kombaiah

CO-INVESTIGATORS:
Colin Judge, INL
Michael Heighes, INL

High-throughput corrosion-fatigue testing capability and expertise will help current reactor lifetime extension and advanced reactor deployment.

Crack initiation and growth in-reactor structural materials under cyclic loading (fatigue) are aggravated by the presence of a corrosive environment. To qualify a material for reactor operation, its susceptibility to environmental degradation must be assessed. Assessing material susceptibility to cracking from irradiation-assisted stress corrosion cracking and corrosion fatigue is a growing concern within the nuclear industry and has been identified by the U.S. Nuclear Regulatory Commission, Electric Power Research Institute, and other industry experts as a knowledge gap of high significance for the nuclear community. INL currently has heavily shielded state-of-the-art irradiation-assisted stress corrosion cracking testing capabilities, and there are similar low activity facilities within the U.S. (e.g., the University of Michigan and the Pacific Northwest National Laboratory). However, no facilities in the U.S. are currently capable of assessing irradiation-assisted corrosion fatigue of materials in a simulated corrosive environment like that of a boiling water reactor or pressurized water reactor. To demonstrate this technique, researchers developed an innovative corrosion-fatigue testing framework capable of testing multiple specimens at a time providing a cost-efficient and accelerated routine to obtain statistical analysis of materials behavior.

Researchers designed and produced a corrosion-fatigue fixture that is capable of multiple specimen tests at a time and applied the direct current potential drop method to detect any crack initiation and propagation that occur during cyclic loading. Furthermore, this proof-of-concept study investigated the susceptibility of a non-irradiated austenitic alloy to crack initiation under cyclic loading and attempted to demonstrate the capability to detect crack initiation using the direct current potential drop method. This new capability advances INL's ability to test structural materials. Next steps include adopting the corrosion-fatigue testing inside hot cells to support the accelerated development of advanced structural materials for nuclear reactor applications.



Corrosion-fatigue fixture during testing.

PRESENTATION:

Song, R., M. McMurtrey, B. Kombaiah, D. Johnson, M. Heighes, P. Xu, and C. Judge, "Environmentally assisted cracking: Theory and practice," *The Minerals, Metals & Materials Society 2022 Annual Meeting & Exhibition*, Anaheim, CA, USA, Feb. 27 – Mar. 3, 2022.

Automated Dependency Graph Partitioning in Multiphysics Object-Oriented Simulation Environment Framework

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PROJECT NUMBER:
20A1054-012

PRINCIPAL INVESTIGATOR:
Robert Carlsen

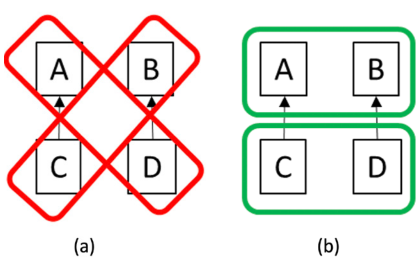
CO-INVESTIGATORS:
Andrew Slaughter, INL
Logan Harbour, INL

Newly developed techniques improve the speed and efficiency of high-fidelity nuclear reactor simulations.

High-fidelity nuclear reactor simulation is critical to ensuring the continued operation of the existing nuclear reactor fleet and the development of next-generation reactors. These simulations are incredibly complex, combining neutronics, heat conduction, solid mechanics, fluid flow, and material science. The MOOSE framework developed by INL has made solving these complex problems possible. This research investigated and developed techniques that can significantly improve the efficiency/speed of existing MOOSE simulations, enable MOOSE to solve problems with greater complexity, and reduce the cognitive overhead and maintenance burden on MOOSE developers and users.

One significant efficiency issue relates to how FEM calculations are performed. These calculations use a mesh of geometric-shaped elements to represent the physical geometry. Performing simulations requires calculating an intricate sequence of quantities associated with mesh locations that all depend on each other. Because many of these calculations can depend on each other, they are currently computed using a set of hard-coded loops over the mesh. For many MOOSE-based applications, this results in unnecessary looping over the mesh with each loop requiring many redundant calculations to be performed. To overcome this inefficiency and other related limitations, a system of annotated dependencies for each of these calculations and an associated partitioning algorithm for automatically executing these calculations were developed and studied. No other FEM or multiphysics codes currently provide similar high-level automated mesh loop generation capability.

Minimizing the total number of mesh loops while honoring all constraints imposed by the needs of each calculation is challenging. Exhaustively checking all possible ways to combine the calculations into loops scales with exponential complexity and is infeasible. While correctly grouping the calculations into loops is relatively easy, doing this in an optimal way is extremely difficult. This problem is effectively a graph partitioning and manipulation problem. Discovering an effective heuristic to maximally combine an arbitrary graph partitioning turned out to be the crux of this research. A heuristic was developed that generates an irreducible set of graph partitions—no further partitions can be combined without first splitting one or more partitions.



***Conflicting consolidation:** When assigning calculations to mesh loops, some combinations are suboptimal because they result in a larger total number of groups. Consider the case of four aggregation calculations: C depends on A, and D depends on B. Combining A and D prevents any further consolidation (a). It would prevent combining C and B (shown) as well as other combinations, such as A and B. However, combining A and B still allows C and D to be combined (b). Much more complicated versions of this problem must be handled by the algorithm. Understanding and solving this phenomenon in all its complex forms was the crux of the research.*

Spectral Observation Convolutional Neural Network

TOTAL APPROVED AMOUNT:
$116,000 over 1 year

PROJECT NUMBER:
20A1054-014

PRINCIPAL INVESTIGATOR:
Ryan Fronk

CO-INVESTIGATOR:
Matthew Anderson, INL

Sophisticated deep learning enhances the value of existing spectroscopic techniques by returning higher-fidelity data and making feature identification possible where it is not presently possible.

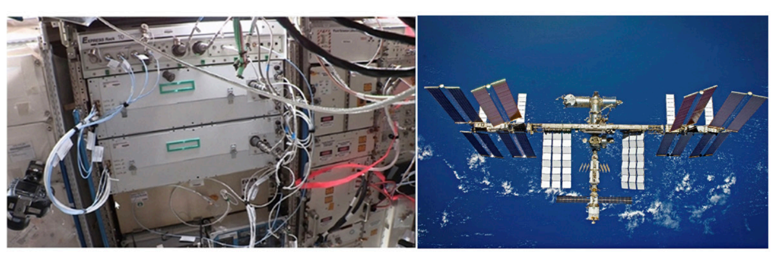
The information that can be unraveled from radiation spectra can enable research and development of novel nuclear fuels and materials. A method to accurately and repeatably analyze spectra is crucial to spectroscopists and researchers across the nuclear industry. The conventional method of identifying and quantifying spectral features from a measurement involves tallying the 'counts' within characteristic energy ranges. This conventional method of analysis introduces uncertainties because features are 'fit' using some form of mathematical fitting to find and then quantify features, which is never perfect. These techniques can also become unreliable under certain circumstances, such as unexpected count rates, elevated background, and complex spectra. Despite substantial research to improve these conventional techniques, the basic method to 'fit' a spectrum to a mathematical model has not changed dramatically for decades.

The SPectral Observation Convolutional neural networK (SPOCK) project set out to explore a method to analyze collected radiation spectra using advanced, scalable deep learning. INL is uniquely equipped to shift the problem of spectroscopy away from a series of mathematical curve fits to a 'recognition' problem using an advanced method of machine learning. INL's pairing of spectroscopic expertise with the newly built and one-of-a-kind Sawtooth supercomputer graphics processing unit cluster are essential to bring this next generation of spectroscopy using machine learning to the larger spectroscopy market.

This concept was developed by training Sawtooth on real-world and simulated gamma-ray spectra. SPOCK analyzed gamma-ray spectra to greater than 99% at the 90% confidence interval. Furthermore, SPOCK was trained, tested, and operated on the International Space Station's Spaceborne Computer-2 supercomputer, returning zero errors over the course of 100 training hours. This demonstrated that SPOCK could perform: (1) autonomously in far-edge, low-wattage computing situations; and (2) in hazardous radiological environments, where interference can cause errors.

TALENT PIPELINE:

- Robyn Hutchins, student at Kansas State University
- Heyley Gatewood, student at Georgia Institute of Technology
- Graeme Holliday, student at University of Idaho



(a) (b)

Spaceborne Computer 2 (a) installed into the International Space Station (b). SPOCK successfully replicated terrestrial training results in a high-radiation environment.

Assessment of Neutron Irradiation Tolerance of Semi-coherent Nano-lamellar Structures

TOTAL APPROVED AMOUNT:
$120,000 over 1 year

PROJECT NUMBER:
20A1054-017

PRINCIPAL INVESTIGATOR:
Cheng Sun

CO-INVESTIGATORS:
Boopathy Kombaiah, INL
Ju Li, Massachusetts Institute of Technology

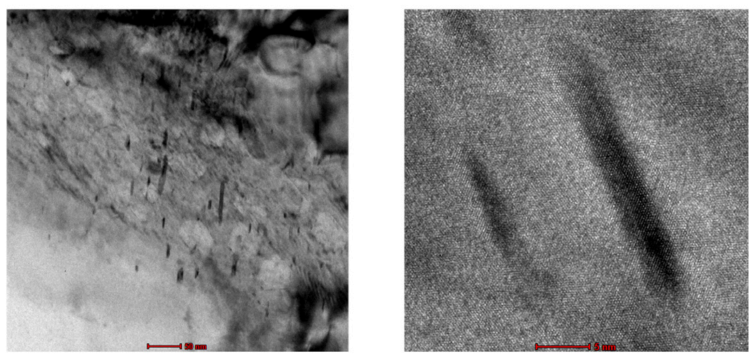
COLLABORATORS:
Center for Materials Elaboration and Structural Studies
Massachusetts Institute of Technology

Fundamental understanding of irradiation response of semi-coherent interfaces provides insights into the design and creation of irradiation-resistant materials.

The objective of this research was to elucidate the role of semi-coherent nano-lamellar structures on the defect evolution and phase stability of intermetallic titanium aluminides under irradiation, with the long-term objective of applying this understanding to design irradiation tolerant materials for advanced nuclear energy systems. In nuclear reactors, irradiation damage significantly degrades the microstructure and mechanical properties of materials, which limits the lifetime of nuclear core components in service. The development of advanced materials that are immune to irradiation damage is one of the great challenges in the design of advanced nuclear reactors. In this project, we studied the irradiation damage of semi-coherent lamellar structures in titanium aluminides under irradiation and demonstrated an approach to enhance the irradiation tolerance of materials via nano-engineering. The success of this project will stimulate broader research interest in the design of irradiation-tolerant materials for harsh environments.

TALENT PIPELINE:

- Haowei Xu, student at Massachusetts Institute of Technology



Planar faults formed in the vicinity of faceted helium gas bubbles in titanium aluminides. The image on the right is a 10x zoom to magnify the planar faults.

Scalable Domain Decomposition Algorithms for Large-scale Monte Carlo Neutron Transport Simulations on Unstructured Mesh

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PROJECT NUMBER:
20A1054-019

PRINCIPAL INVESTIGATOR:
Guillaume Giudicelli

CO-INVESTIGATOR:
Logan Harbour, INL

Multiphysics simulations support the advancement of clean energy solutions.

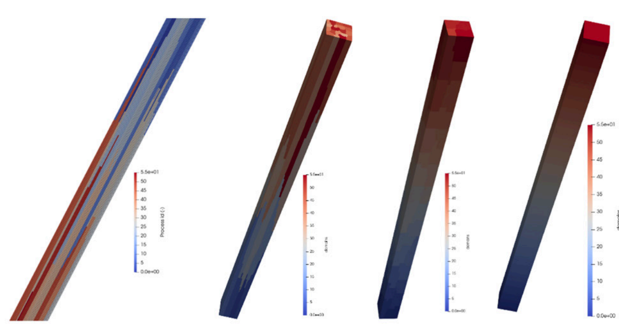
The main physics involved in nuclear reactors are neutronics, thermal-hydraulics, and fuel thermo-mechanics. Multiphysics simulations are only as accurate as the models used for each of these physics. Though Monte Carlo neutral particle transport methods are widely regarded as the gold-standard for neutronics simulations, they were long thought impractical for most types of nuclear reactor analysis outside of shielding calculations.

However, with the current drive towards higher-fidelity nuclear reactor analyses, as well as the growing interest in small modular reactors, the method has become competitive with traditional deterministic transport algorithms for the same level of accuracy. Before such analysis can be practical, several algorithmic challenges must be addressed, particularly with regards to the memory requirements of the method. The most common approach to these issues is to perform a domain decomposition to share the memory burden among numerous nodes of a computing cluster. To date, most Monte Carlo codes do not offer domain decomposition as existing implementations lack practicality, performance, or scalability. This limits their use for large-scale simulations, for example of nuclear reactor cores in neutron transport simulations or full tokamak models in shielding calculations. While most Monte Carlo codes use simplified geometric models known as Constructive Solid Geometry, support for computer-aided design models and unstructured mesh in Monte Carlo codes is increasingly common and required to model complex geometries of advanced reactors or fusion devices. The memory requirements of an unstructured mesh exacerbate the memory issues of the Monte Carlo method and, therefore, the need for domain decomposition.

This project first involved the development of a Monte Carlo neutron transport application, MaCaw, for the MOOSE framework, based on physics routines from OpenMC and ray-tracing routines developed in MOOSE for the method of characteristics transport. The application supports both shared memory and distributed memory parallelism. The latter is achieved using domain decomposition. In this situation, the tallies are also domain decomposed, splitting the memory cost of large simulations among all processes. This new application is being tested on a light water reactor full core benchmark. In its final phase, this project addressed the issue of communication of particles between domains and the issue of load balancing to improve scalability.

TALENT PIPELINE:

- Robert Crowder, student at University of Tennessee Knoxville



Fuel pin partitioners available in the MOOSE ecosystem for Monte Carlo particle transport simulations. From left to right: equal volume, default (based on element numbering), minimization of domain surfaces, regular axial grid.

Reactor Core Isolation Cooling System Model Development and Implementation in the Reactor Excursion and Leak Analysis Program

TOTAL APPROVED AMOUNT:
$60,000 over 1 year

PROJECT NUMBER:
21A1057-006

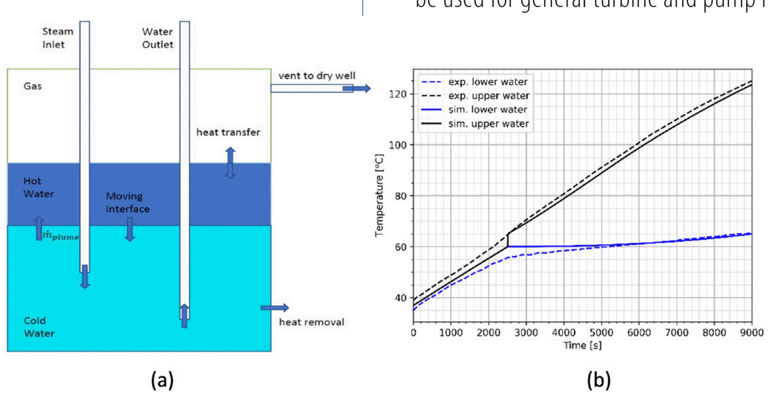
PRINCIPAL INVESTIGATOR:
Jack Cavaluzzi

CO-INVESTIGATOR:
David Andrs, INL

Mechanistic component models improve boiling water reactor risk assessments.

The reactor core isolation cooling (RCIC) system is an emergency safety system used in over 20 U.S. boiling water reactors. The coupled system can cool the core with little to no outside power and is composed of a Terry steam turbine, a pump, and the containment wetwell. The 2011 accident at Fukushima Daiichi highlighted the importance of the RCIC system, which provided core cooling to Unit 2 for 70 hours during an extended station blackout. Current systems-level codes model the RCIC system with generic models that do not consider RCIC-specific phenomenon and therefore have limited ability to correctly simulate event progression during RCIC system operation. The goal of this project was to develop physics-based models of an integral RCIC system and incorporate the models into the Reactor Excursion and Leak Analysis Program (RELAP)-7 code.

This project resulted in new RELAP-7 component models that can easily and accurately simulate long-term operation of a complete RCIC system. An original wetwell model was implemented in RELAP-7. This model captures buoyancy-induced thermal stratification effects due to injection of the Terry turbine exhaust steam. This phenomenon is not accounted for in current systems-level analysis codes, yet the pressure suppression capacity of the wetwell is largely influenced by the energy redistribution due to RCIC turbine steam exhaust. Original one- and two-phase turbine models were developed and implemented in RELAP-7. The new wetwell, one-phase turbine, and existing one-phase pump components were coupled to simulate a complete RCIC system at normal operating conditions. The two-phase turbine component was designed to have performance degradation as a function of incoming liquid water content. The two-phase turbine was coupled to the one-phase pump to demonstrate the "self-regulating" mode that was observed at Fukushima Daiichi Unit 2. These new component additions enhance modeling capability and are valid for a wider range of RCIC operation space than current systems-level analysis codes. The mechanistic component models allow simulation of long-term RCIC performance, which can improve boiling water reactor plant guidelines and risk assessments. The improved turbomachinery components can also be used for general turbine and pump modeling.



(a) Two-zone RCIC schematic. (b) Stratified wetwell simulation results compared to experimental data from Texas A&M Nuclear Heat Transfer Systems Laboratory.

TALENT PIPELINE:

- Jack Cavaluzzi, student at Texas A&M University

PUBLICATION:

Cavaluzzi, J., D. Andrs, and K. V. Kirkland, "Two-zone stratified wetwell model development and implementation for RELAP-7," Annals of Nuclear Energy 164 (2021) 108592.

Closed-form Solution for the Movement of Hydrogen in Nuclear Thermal Propulsion Reactor Fuels during Transient Operation

TOTAL APPROVED AMOUNT:
$275,000 over 1 year

PROJECT NUMBER:
21P1063-001

PRINCIPAL INVESTIGATOR:
Stephen Johnson

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

COLLABORATOR:
Center for Space Nuclear Research

Modeling the fundamental physics related to nuclear thermal propulsion makes space travel faster and safer.

Future human exploration of Mars and the outer solar system will require the use of nuclear energy to reduce travel time and the exposure of the crew to energetic protons (i.e., cosmic rays and solar flares) in space. The propulsion concept that has been developed the most is nuclear thermal propulsion, a very high temperature reactor in which hydrogen is heated from 20 K to about 2700 K before exiting through a nozzle at about 10 km/s, producing a specific impulse of ~900 s, nearly double that of conventional rockets. Since 2018, the U.S. National Aeronautics and Space Administration has been sponsoring renewed research, some at INL, to develop nuclear fuels for such very high temperature, high specific impulse reactors.

The main project used data and correlations from the nuclear engine for rocket vehicle application program to develop a model for the solubility of hydrogen in the tungsten-rhenium coolant tubes as a function of temperature and time, then to use that model to estimate the pressure of hydrogen in bubbles along grain boundaries in the coolant flow tubes as a function of time and of location within the reactor during the shutdown transient. The goal of the model is to provide criteria for the ramp-down of hydrogen coolant flow needed to prevent decohesion of grains in the coolant tubes. If the hydrogen coolant flow remains too high following control drum rotation, then the reactor will cool rapidly, trapping hydrogen along the grain boundaries in the tubes and failing the tubes. If the hydrogen coolant flow decreases abruptly following control drum rotation, then there will be insufficient transport of the reactor decay heat and the fuel, already operating near its melting point, may overheat and deform. Thus, it is critical to know the boundaries for safe shutdown of the nuclear thermal propulsion reactor.

A second, smaller project was to develop a model for the highbay room at the Materials and Fuels Complex where the fueled Multi-Mission Radioisotope Thermoelectric Generator is loaded into its shipping cask. The purpose of this model is to allow the experimental measurement of the neutron and gamma fluxes and spectra produced by the Multi-Mission Radioisotope Thermoelectric Generator within the concrete-walled highbay and the extrapolation of those measurements to the materials and configuration of the spacecraft and surroundings during its mission. This capability is particularly important where there are sensitive instruments, such as optical spectrometers or pulsed neutron sources on a drone in a planetary atmosphere such as is being proposed for the Dragonfly mission to Titan, the largest moon of Saturn.

TALENT PIPELINE:

- Benjamin Masters, student at Virginia Polytechnic Institute and State University
- Berenice Sosa Aispuro, student at Idaho State University
- Chaitee Godbole, student at North Carolina State University
- Emory Colvin, student at Oregon State University
- Kean Martinic, student at Idaho State University
- Matthew Simmers, student at Virginia Polytechnic Institute and State University
- Nicholas Morris, student at University of Alabama, Huntsville
- Rahma Ali, student at Stanford University
- Teyen Widdicombe, student at University of Idaho
- Zane Emery, student at Cornell University

PUBLICATION:

Widdicombe, T., and R. Borrelli, "MCNP modeling of radiation effects of the Dragonfly mission's radioisotope thermoelectric generator (RTG) on Titan," *Acta Astronautica* 183(7) (2021) 363–373.

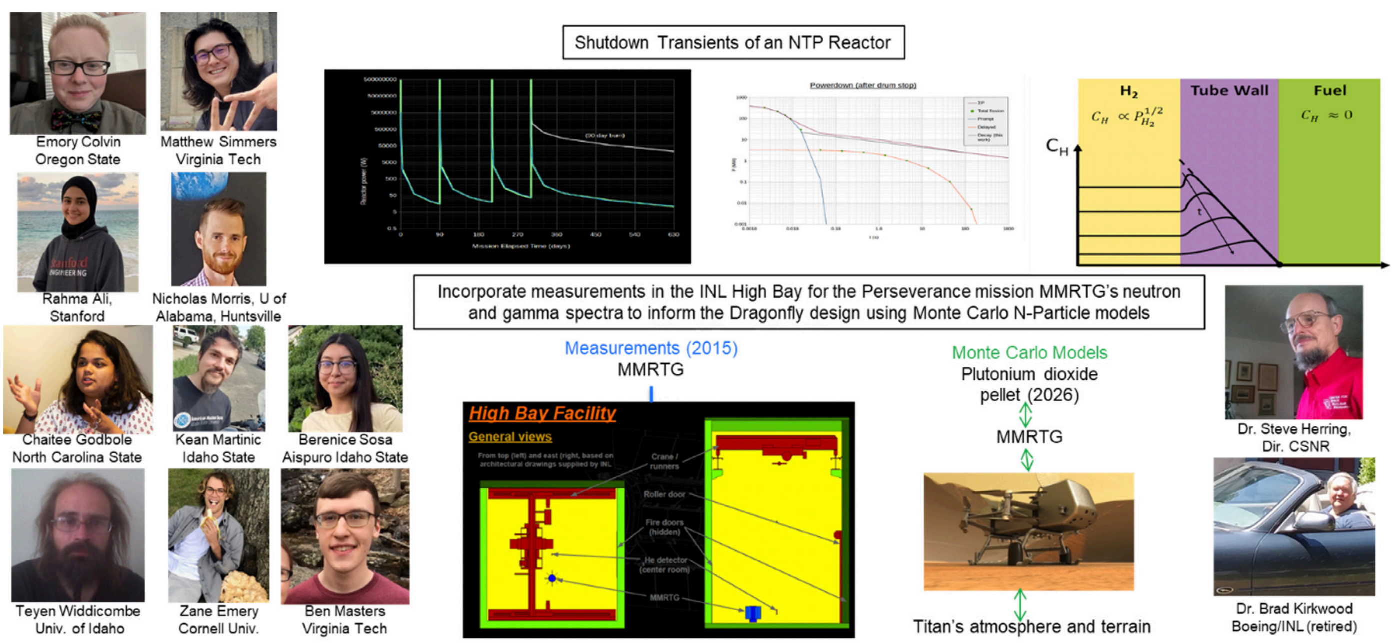
PRESENTATIONS:

Widdicombe, T., A. Grieve, S. Herring, and B. Kirkwood, "Design of NERVA-derived HALEU reactor for nuclear thermal propulsion," *Nuclear and Emerging Technologies for Space 2021*, Virtual, April 26–30, 2021.

Colvin, E., "Optimization of plutonium-238 production in the Advanced Test Reactor," *Nuclear and Emerging Technologies for Space 2021*, Virtual, April 26–30, 2021.

Rhodes, J., E. Colvin, T. Widdicombe, and B. Kirkwood, "Mitigation of 208Tl gamma dose from 236Pu decay chain via chemical removal of 232U," *Nuclear and Emerging Technologies for Space 2021*, Virtual, April 26–30, 2021.

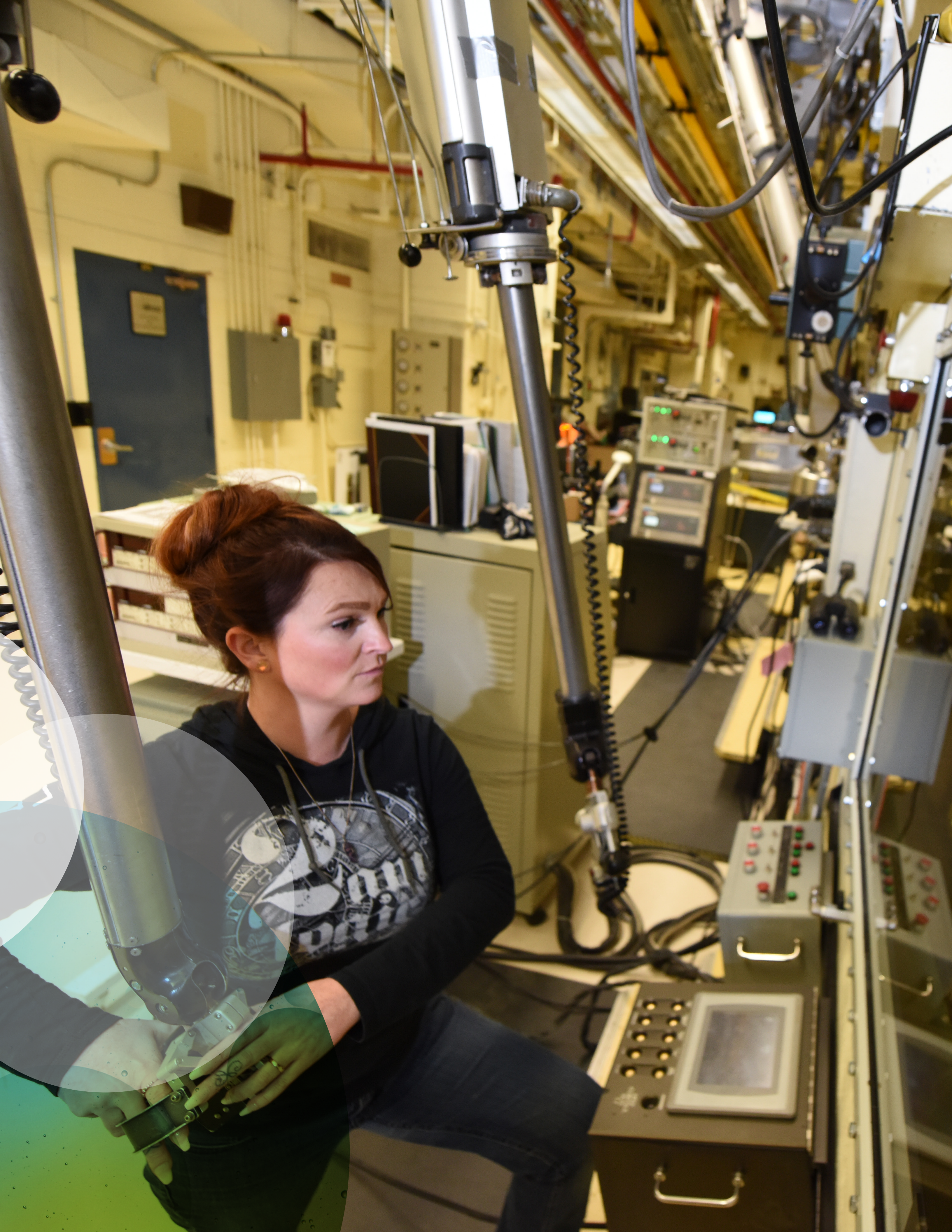
Kajihara, T., C. Godbole, and J. Magnusson, "Neutronic analysis of the submersion subcritical safe space (S4) reactor using reduced enrichment uranium fuel (35949)," *Nuclear and Emerging Technologies for Space 2021*, Virtual, April 26–30, 2021.



The 2021 Center for Space Nuclear Research Summer Program fellows participated virtually.

INTEGRATED FUEL CYCLE SOLUTIONS

Sustainability of nuclear energy systems requires addressing economic, environmental, and social dimensions and is key to the acceptance of nuclear energy as part of the solution to climate change. Advanced reactor development and deployment presents an opportunity to challenge the traditional fuel cycle approach and develop disruptive, integrated fuel cycle solutions. Both special nuclear material and new waste generation require a fuel cycle with inherent process transparency that reduces the risk of nuclear proliferation. INL is focused on developing fuel cycle technologies that incorporate safeguards by design for the effective and efficient monitoring and verification of nuclear materials throughout the fuel cycle. INL supports proliferation risk reduction in the integrated fuel cycle through research that demonstrates simplified used nuclear fuel recycling processes, develops and demonstrates real-time interrogation of used nuclear fuel treatment processes, and demonstrates the direct chemical and physical immobilization options for used nuclear fuel.



Expanding Photonuclear Data via High-Energy Linear Accelerator Coupled with Real-time Gaseous Mass Separation

TOTAL APPROVED AMOUNT:
$1,537,750 over 3 years

PROJECT NUMBER:
18P37-031

PRINCIPAL INVESTIGATOR:
Kevin Carney

CO-INVESTIGATOR:
Jared Horkley, INL

COLLABORATOR:
Idaho State University

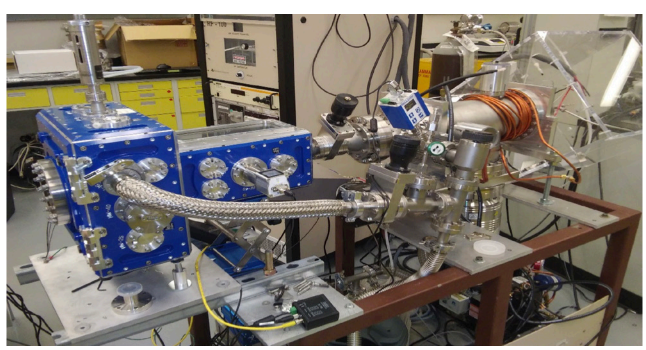
Novel study enables future research and computational science in photonuclear reactions for isotope production for national security, medical, and astrophysics applications.

Accelerator-driven photonuclear reactions are being used more frequently to support scientific discovery, nuclear security, and medical and industrial isotope production. Inadequacies in the abundance and accuracy of photonuclear data, specifically reaction cross-sections, hamper accelerator use. This research project proposed to improve the availability of measured photonuclear cross-section data and potentially nuclear data for gamma spectrometry by integrating technologies for isotope separation with isotope production. Coupling a bremsstrahlung linear accelerator with an isotope separator has the potential to expand the library of measured nuclear data associated with energy-dependent photonuclear reactions, particularly for low-yield reactions involving short-lived isotopes. This tandem technique enables the isolation, observation, and collection of fission and activation products for many isotopes with half-lives under one minute. The rapid separation of decay-chains by mass on time frames from seconds to hours will remove spectral gamma-ray congestion and improve spectral assignments and data for relative production yields for gamma induced fission, proton and neutron reactions as a function of bremsstrahlung excitation energy. Bremsstrahlung irradiations provide an alternate method to investigate the production of isotopes via proton accelerator-induced nuclear reactions with significantly less risk since the need for thin window targets are not required.

This project had several components: (1) confirming that a low-resolution, low-cost isotope separator can adequately provide: (a) the resolution for unit mass separation of isotopes; (b) real-time detection of isotope abundances; and (c) adequately separate and collect isotopes to simplify gamma spectra from radioactive isotopes that are measured to determine reaction cross-sections; (2) reconstituting the capability to calculate cross-sections from measured reaction yield data; and (3) demonstrating the isolation of activated isotopes and capability to obtain cross-section data on minor photonuclear reaction products.

TALENT PIPELINE:

- Ariana Foley, student at Oregon State University
- Daryl Giglio, student at The Ohio State University
- Thibaut Lecrevain, INL
  Glenn T. Seaborg Distinguished Postdoc



Isotope collection system with simultaneous detection and collection (10 isotopes) and beam profile monitoring.

Liquid Scintillation Alpha Spectrometry for Nuclear Non-proliferation and Nuclear Forensics

TOTAL APPROVED AMOUNT:
$735,000 over 3 years

PROJECT NUMBER:
19A39-020

PRINCIPAL INVESTIGATOR:
David Chichester

CO-INVESTIGATORS:
James Johnson, INL
Mathew Snow, INL
Scott Thompson, INL
Scott Watson, INL
Thomas Holschuh, INL
Guy Backstrom, Radiological and Environmental Sciences Laboratory

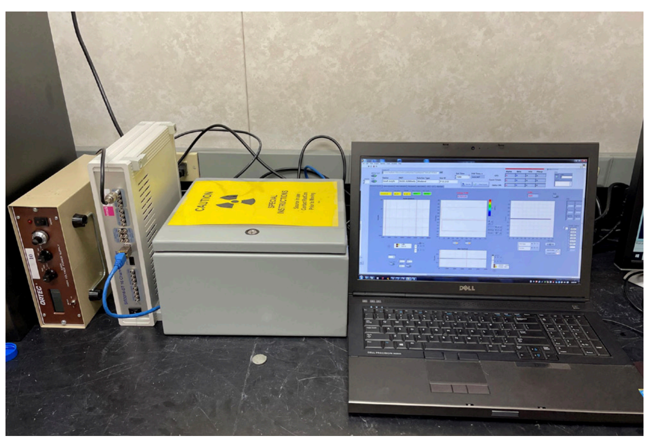
Spectrometer advances improve actinide isotope detection in environmental samples for nuclear non-proliferation, nuclear energy, environmental monitoring, and other applications.

Researchers hypothesized that pulse-shape discrimination alpha liquid scintillation spectroscopy employing current state-of-the art electronics and waveform signal analysis methods can achieve milli-becquerel alpha emitter detection limits, and that these methods can greatly improve the ability to detect and quantify actinide isotopes in environmental samples for nuclear non-proliferation applications, nuclear energy applications, environmental monitoring, and other applications. A prototype alpha spectrometry tool was developed that is suitable for laboratory or field work and can assay liquid solutions or filter paper placed in a solution and determine the isotopic content of the alpha-emitting nuclides. Key innovations in this approach include: (1) the use of advanced digital signal processing methods and equipment in place of analog electronics; (2) the use of robust miniaturized silicon photomultipliers in place of vacuum-tube photomultipliers; and (3) the use of new chemical methods that do not generate hazardous mixed waste.

As a result of this proof-of-principle research, an operator can mix an analyte solution with a pre-prepared liquid scintillation solution and then place the mixture into a measurement device the size of a small box. The prototype system includes hardware and automated software to perform the analysis. Results are reported that include the alpha-emitting isotope activity in units of becquerel, including uncertainty.

TALENT PIPELINE:

- Creighton King, student at Washington State University
- Jessica Ward, student at University of Idaho



Pulse-shape discrimination alpha liquid scintillation spectroscopy enables field-ready alpha emitter detection at milli-becquerel levels.

Dissolution Phenomena of Used Nuclear Oxide Fuel in Molten Salt Systems

TOTAL APPROVED AMOUNT:
$1,127,500 over 3 years

PROJECT NUMBER:
19A39-059

PRINCIPAL INVESTIGATOR:
Steven Herrmann

CO-INVESTIGATORS:
Steven Frank, INL
Brian Westphal, INL
Haiyan Zhao, University of Idaho

Novel chemistry facilitates used nuclear fuel reuse in fast reactor systems where transuranium constituents can be burned.

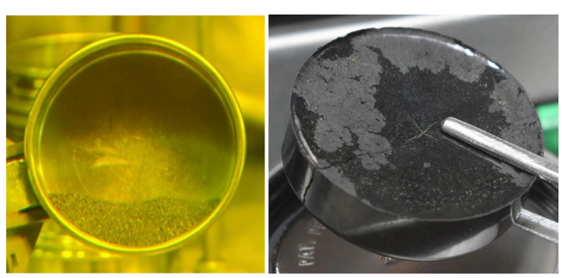
The objective of this research was to demonstrate and characterize a novel technique for dissolution of used nuclear oxide fuel constituents in select molten salt systems to facilitate their reuse in fast reactor systems. Used nuclear oxide fuel constituents were converted into molten chloride systems for potential use as: (1) molten chloride salt reactor fuel; or (2) a head-end step to group actinide metal recovery via electrochemisry. Both conversion options facilitate used nuclear oxide fuel reuse in fast reactor systems, where transuranium constituents can be burned as a metal fuel or a molten chloride salt fuel. Dissolution of used nuclear oxide fuel constituents were demonstrated by immersing fuel particulate into a molten salt system containing a base chloride salt (e.g., alkali metal chlorides) and a chlorinating agent (e.g., uranium trichloride). Applying an electric current to some oxide fuel assisted dissolution by promoting conversion of transuranium, lanthanide, alkaline earth, and alkali oxide species from the fuel into chlorides. Noble metal fission products were not converted to chlorides in this system and were consequently removed with uranium oxide in an insoluble solid form. Lithium chloride, sodium chloride, potassium chloride, or combinations thereof, were used as the base salts in this system. A lithium-potassium chloride base salt best served subsequent removal of group actinide metals while sodium chloride was better suited for a molten chloride salt reactor fuel. Oxide fuel preconditioning and in situ reducing conditions along with elevated temperature and uranium trichloride concentrations were the primary parameters promoting used nuclear oxide fuel constituent dissolution in accordance with identified reaction mechanisms during this demonstration, where 90 – 99+% of transuranium and reactive fission product constituents dissolved into the subject molten salt systems. High-purity uranium trichloride was synthesized and characterized in this study to support the oxide fuel dissolution.

TALENT PIPELINE:

- Steven Herrmann, student at University of Idaho

PUBLICATION:

Herrmann, S. D., B. R. Westphal, S. X. Li, and H. Zhao, "Parametric study of used nuclear oxide fuel constituent dissolution in molten $LiCl\text{-}KCl\text{-}UCl_3$," *Nuclear Technology* (2021) Ahead-of-Print. Available at: https://doi.org/10.1080/00295450.2021.1973180.



(a) (b)

(a) Declad used nuclear oxide fuel. (b) Synthesized lithium-potassium-uranium chloride salt ingot.

Constituent Redistribution in Nuclear Fuels

TOTAL APPROVED AMOUNT:
$1,065,000 over 3 years

PROJECT NUMBER:
19A39-100

PRINCIPAL INVESTIGATOR:
Michael Benson

CO-INVESTIGATORS:
James King, INL
Pavel Medvedev, INL
Jinsuo Zhang, Virginia Tech

Simulating nuclear reactor temperature conditions enables faster and lower-cost discovery of new nuclear fuels.

The objective of this research was to develop and demonstrate an apparatus capable of generating a temperature gradient in a nuclear fuel pin that can be applied to any existing or novel nuclear fuel pin composition. Out-of-pile testing is a crucial component of understanding fuel behavior for both existing and new fuel compositions. Compared with a reactor experiment, an out-of-pile experiment takes weeks or months instead of years and does not have the post-irradiation fuel handling requirements. To be as effective as possible, out-of-pile testing needs to reproduce reactor conditions as closely as possible. To this end, control of the temperature gradient has been incorporated into the experiments. This allows for the simulation of the temperature gradient found in nuclear fuel during irradiation and simulation of off-normal events. This research provides significant cost-savings over traditional reactor experiments and provides necessary information to the nuclear fuel modeling effort.

Different compositions of uranium-molybdenum-titanium-zirconium (U-Mo-Ti-Zr) fuel alloys were characterized as part of this effort. This composition was chosen for multiple reasons, such as the fact that molybdenum and zirconium have significantly different redistribution behavior and the potential to maintain gamma (γ) phase at the temperatures found in a fast reactor. As an example, the alloy U-1.5Mo-1.5Ti-7.0Zr (U-MT7Z) undergoes a phase transition from the alpha (α) phase to the γ phase on annealing above 675˚C. The phase transitions were determined by DSC, X-ray diffraction, and scanning electron microscopy. Understanding these phase transitions is fundamental to understanding metal alloy fuel behavior.

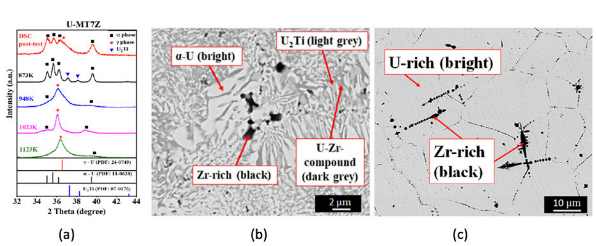
TALENT PIPELINE:

- Weiqian Zhou, student at Virginia Polytechnic Institute and State University
- Huali Wu, postdoc at Virginia Polytechnic Institute and State University

PUBLICATIONS:

Zhuo, W., Y. Xie, M. T. Benson, J. B. Ge, R. D. Mariani, and J. Zhang, "XRD and SEM/EDS characterization of two quaternary fuel alloys (U-2.5Mo-2.5Ti-5.0Zr and U-1.5Mo-1.5Ti-7.0Zr in wt. %) for fast reactors," *Materials Characterization* 170 (2020) 110696.

Zhuo, W., H. Wu, Y. Xie, M. T. Benson, and J. Zhang, "Solid-state phase transitions of two quaternary metallic fuel alloys (U-2.5Mo-2.5Ti-5.0Zr and U-1.5Mo-1.5Ti-7.0Zr in wt. %)," *Journal of Nuclear Materials* 555 (2021) 153134.



(a) X-ray diffraction results for U-MT7Z alloy, as quenched from high temperatures. (b) Scanning electron microscopy image of U-MT7Z annealed and quenched from 600°C. (c) Scanning electron microscopy image of U-MT7Z annealed and quenched from 675°C.

Probing Microstructure-induced Swelling and Thermal Property Changes in Ion-irradiated Oxide Fuels using Laser-generated Surface Acoustic Waves

TOTAL APPROVED AMOUNT:
$210,000 over 3 years

PROJECT NUMBER:
19P43-012

PRINCIPAL INVESTIGATOR:
Amey Khanolkar

CO-INVESTIGATORS:
David Hurley, INL
Zilong Hua, INL

COLLABORATORS:
Air Force Research Laboratory
Texas A&M University
The Ohio State University

Advancing fundamental understandings of the role of radiation-induced damage on the thermal transport and mechanical properties of nuclear fuels supports the efficient, reliable, and safe operation of existing and future nuclear power plants.

Laser-based characterization methods have emerged as promising non-contact, non-destructive tools to measure the evolution of microstructure-induced material property changes. Moreover, laser methods offer the promise of in situ characterization while the material is being irradiated. Although recent advances have been made in laser-based property evaluation of irradiated metallic material, little effort has been dedicated to corresponding measurements in non-metallic materials that include semiconductors and ceramics. This research improved the understanding of radiation-induced changes to material microstructure and on the thermal and elastic properties of ceramic nuclear fuels. Low-dose ion irradiation was used to seed specific defect types in the microstructure of thorium oxide (ThO_2) fuel samples to simulate and systematically study the effects of fission fragment damage and lattice swelling on fuel material performance. Transient grating spectroscopy (TGS) was used to simultaneously measure thermal diffusivity and elastic properties at the micrometer length-scale in pristine (unirradiated) and ion-irradiated thorium oxide. TGS uses two crossed laser beams derived from a pulsed nanosecond laser to create a spatially sinusoidal temperature profile (thermal grating) in the sample. Rapid thermal expansion of the sample in the peaks of this thermal grating launches counter-propagating surface acoustic waves with wavelength defined by the thermal grating spacing. The dynamics of the thermal and acoustic response are monitored via the time-dependent diffraction of a continuous wave laser with high beam quality.

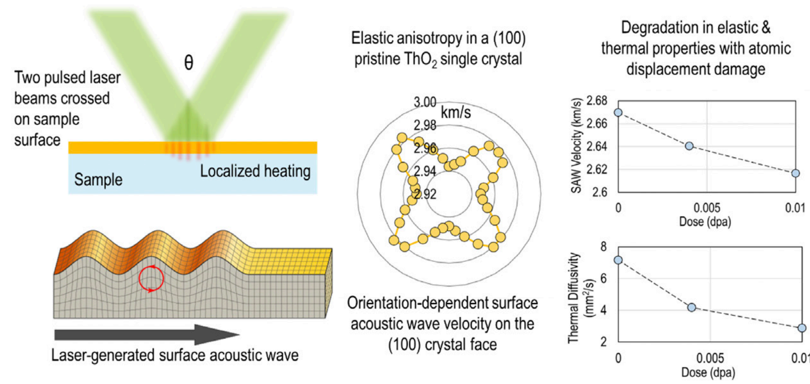
Results from this study demonstrate the sensitivity of the laser-generated surface acoustic waves in detecting radiation-induced elastic and thermal property changes in ceramic energy materials and provide insight into the role of early-stage lattice defects (predominantly point defects) on phonon-mediated thermal transport and elastic properties. This research provides foundational work to validate the influence of atomic-level lattice defects in fuel performance codes. Future studies can use the TGS technique to measure the phonon mean free path at low temperatures. The multi-modal nature of the laser-based TGS technique coupled with the experimental methodology being developed in this project can accelerate new nuclear fuel materials qualification for advanced reactor concepts by reducing the number of iterative measurement steps. In addition to studying radiation effects of thermal and elastic properties of ceramic fuels, the experimental methodology can be applied to intra-granular measurements in complex metal alloys, as well as investigating spatially-confined temperature- and pressure-dependent microstructural phenomena in material systems of interest to the broader condensed matter physics and materials science communities beyond the realm of nuclear fuels.

TALENT PIPELINE:

- Amey Khanolkar, INL
  Russell L. Heath Distinguished Postdoc

PRESENTATION:

Khanolkar, A., Z. Hua, C. A. Dennett, M. Khafizov, J. M. Mann, and D. H. Hurley, "The influence of radiation-induced microstructural defects on the optical and elastic properties of ceramic nuclear fuels," *The Minerals, Metals & Materials Society 2022 Annual Meeting & Exhibition*, Anaheim, CA, USA, Feb. 27 – Mar. 3, 2022.



The transient thermal grating induced by the crossed pulsed pump laser enables non-contact, non-destructive measurement of microstructure-induced material property changes.

Electronic and Thermodynamic Properties of Selected Uranium Compounds at High Magnetic Fields

TOTAL APPROVED AMOUNT:
$235,000 over 3 years

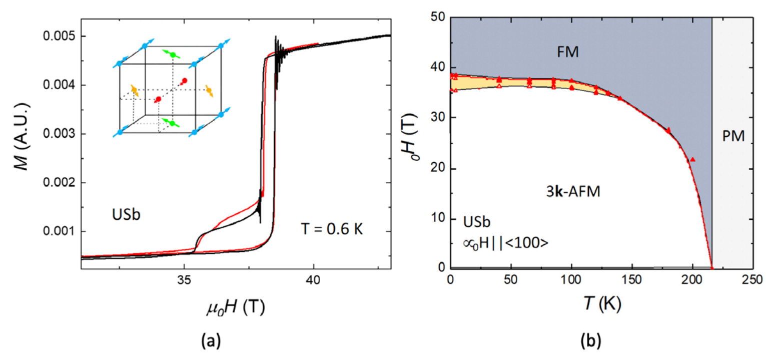
PROJECT NUMBER:
19P43-013

PRINCIPAL INVESTIGATOR:
Xiaxin Ding

CO-INVESTIGATOR:
Krzysztof Gofryk, INL

This novel research resulted in a better understanding of electronic, thermodynamic, and elastic properties in actinide materials, which are important in developing new advanced (metallic) nuclear fuels.

Thermodynamic and electronic behaviors that involve 5*f* electrons in the actinide materials exhibit a bewildering diversity that is difficult to fit within any conventional framework. This project focused on fundamental understanding of thermodynamic and transport properties of selected 5*f*- and 5*d*-electron materials that show both strong electronic correlations and strong spin-phonon interactions. Despite the large experimental and theoretical works carried out, the nature of the *f*-electrons and their coupling to lattice vibration and magnetic fluctuations are still unclear. This project included systematic studies of uranium monoantimonide (USb) and vanadium telluride. To better understand the interplay of many-body physics and other degrees of freedom, such as strong electronic correlations, magnetism, and structural distortions, researchers performed detailed magnetic, transport, and thermodynamic, studies of these materials at low temperatures and very high magnetic fields (both pulsed and continuous). This new fundamental research related to correlated materials at low temperatures and high magnetic fields led to a better understanding of the Kondo effect in vanadium telluride and to the unveiling, for the first time, a magnetic phase diagram for USb. These new results will increase our knowledge of electronic correlations in 5*f*-electron materials, which is of interest to the wide condensed matter physics community.



(a) The magnetic field dependence of magnetization of USb crystal measured at 0.6 K and in pulsed magnetic fields up to 65 T. The oscillation in M(H) curve comes from the Gibbs effect and suggests the first order nature of the transition. Inset: 3k magnetic structure of USb. (b) A magnetic phase diagram of USb.

TALENT PIPELINE:

- Xiaxin Ding, INL
  Glenn T. Seaborg Distinguished Postdoc

PUBLICATIONS:

Ding, X., J. Xing, G. Li, L. Balicas, K. Gofryk, and H.-H. Wen, "Crossover from Kondo to Fermi-liquid behavior induced by high magnetic field in 1T—VTe_2 single crystals," *Physical Review* B 103 (2021) 125115.

Willis, X., X. Ding, J. Singleton, and F. F. Balakirev, "Cryogenic goniometer for measurements in pulsed magnetic fields fabricated via additive manufacturing technique," *Review of Scientific Instruments* 91 (2020) 036102.

Sharma, M., A. Yangui, A. Lusson, K. Boukheddaden, X. Ding, M.-H. Du, K. Gofryk, and B. Saparov, "Additive-assisted synthesis and optoelectronic properties of $(CH_3NH_3)_4Bi_6I_{22}$," *Inorganic Chemistry Frontiers* 7 (2020) 1564–1572.

PRESENTATIONS:

Ding, X., K. Shrestha, D. Antonio, F. Weickert, M. Jaime, N. Harrison, M. B. Salamon, T. Durakiewicz, J. L. Smith, and K. Gofryk, "Magnetic phase diagram of uranium monoantimonide single crystals," 74th *Calorimetry Conference*, Santa Fe, NM, USA, July 28 – Aug. 2, 2019.

Ding, X., "Physical properties of actinide materials under extreme conditions," *Annual Review Glenn T. Seaborg Institute*, Idaho National Laboratory, Idaho Falls, ID, USA, Oct. 24, 2019.

Ding, X., "Magnetic and electronic properties of correlated materials," *Annual Review Glenn T. Seaborg Institute*, Idaho National Laboratory, Idaho Falls, ID, USA, Nov. 11, 2020.

Production of Scandium-47 for Potential Use as a Radiotheragnostic

TOTAL APPROVED AMOUNT:
$1,060,000 over 3 years

PROJECT NUMBER:
19P45-028

PRINCIPAL INVESTIGATOR:
Mathew Snow

CO-INVESTIGATORS:
Ariana Foley, INL
Jessica Ward, INL
Tara Mastren, University of Utah

COLLABORATOR:
Idaho State University

Purified radioisotopes enable breakthroughs for nuclear energy, medical treatment, and national security.

Prior to this research, producing large quantities and high purities of critical isotopes and short-lived, low-yield fission products either could not be done or required extremely expensive materials that have limited stockpiles available worldwide. INL researchers developed an innovative approach using high-energy photons that offers high potential for on-site and less-expensive production due to the broader availability of linear accelerators at government laboratories, research institutions, and medical facilities around the world. Many of these new pathways can be designed to use readily available, low cost, high-purity target materials that produce more pure isotopes of interest when irradiated, resulting in simplified purification processes that reduce the time to purify the isotopes and minimize radiation exposure. For example, the new approach developed through this research to produce scandium-47 utilizes natural vanadium, which is relatively inexpensive and virtually unlimited in supply as compared to other techniques. Using a natural vanadium target also avoids harmful gamma radiation from the byproducts produced during scandium-47 production.

Researchers will expand into synthesizing heavy lanthanide fission products relevant to the strategic materials needed for energy research and non-proliferation treaty verification. The production pathways developed through this research may provide the only currently viable approaches for access to these rare isotopes.

Researchers at INL have developed and demonstrated many new, tunable chemical separation approaches that are extremely low cost, use commercially available materials, and result in significant breakthroughs in separation timelines. For example, one approach enables isolation of many key fission product isotopes from highly radioactive samples in under two hours, instead of one week or more using traditional techniques. This work has delivered significant improvement over current techniques in terms of time, chemical recovery, separation, and gamma emissions. The rapid nature, purity, and safety enhancements of the overall processes represents an opportunity for U.S. isotope manufacturers to offer on-demand and on-site rare isotope products and services in the future.

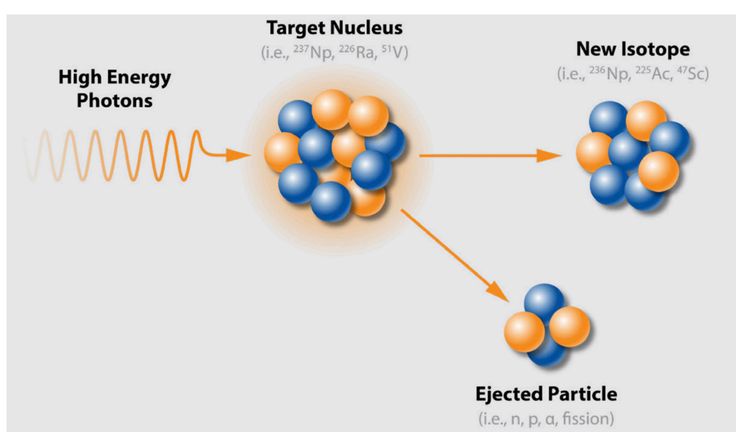


Illustration of the photonuclear reaction process. High-energy photons are impinged on a target material (e.g., neptunium-237, radon-226, vanadium-51), resulting in the ejection of a portion of that nucleus to produce new isotopes (e.g., neptunium-236, actinium-225, scandium-47) of high value to a range of scientific applications.

TALENT PIPELINE:

- Ariana Foley, student at Oregon State University
- Jessica Ward, student at University of Idaho
- Robert Lusk, student at Washington State University

AWARD:

LDRD Highlights at the National Laboratories, 2021, pp. 26–27.

PUBLICATIONS:

Snow, M. S., J. Ward, B. Bucher, J. T. Cooper, M. T. Kinlaw, E. Cardenas, J. Horkley, H. Town, M. Finck, and K. Carney, "Rapid separation of photofissioned uranium products via a single-pass multiplexed chromatographic fission product separation system," *Analytical Chemistry* 93(8) (2021) 3770–3777.

Snow, M. S., A. Foley, J. L. Ward, M. T. Kinlaw, J. Stoner, and K. P. Carney, "High purity 47Sc production using high-energy photons and natural vanadium targets," *Applied Radiation and Isotopes* 178 (2021) 109934.

Ward, J. L., B. Bucher, K. Carney, and M. Snow, "Exploring lanthanide separations using Eichrom's Ln Resin and low-pressure liquid chromatography," *Journal of Radioanalytical and Nuclear Chemistry* 327 (2021) 307–316.

Snow, M., and J. Ward, "Fundamental distribution coefficient data and separations using Eichrom extraction chromatographic resins," *Journal of Chromatography A* 1620 (2020) 460833.

INTELLECTUAL PROPERTY:

Snow, M. S., and M.T. Kinlaw, "Methods of producing enriched scandium-47, and related systems and apparatuses," Patent Cooperation Treaty Application PCT/US20/31422, (May 5, 2020).

Snow, M. S., J. Ward, A. Foley, M. Kinlaw, J. Cooper, and K. Carney, "Methods for producing radionuclides using accelerator Bremsstrahlung irradiation and rapid chromatographic isotope separation," U.S. Patent Application No. 63/203,858 (August 2, 2021).

Proof of Principle Experiments for a Pyrochemical Approach to Treatment of Used Advanced Test Reactor Fuel

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PROJECT NUMBER:
20A1047-039

PRINCIPAL INVESTIGATOR:
Steven Herrmann

CO-INVESTIGATORS:
Michael Patterson, INL
Steven Frank, INL

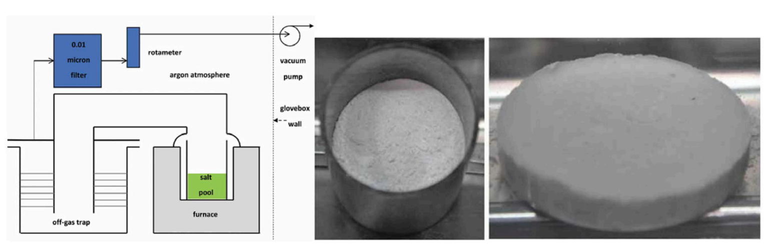
This research expands knowledge in molten salt technologies and benefits innovative fuel cycle science.

The goal of this research was to substantiate a novel pyrochemical process applicable to treatment of used Advanced Test Reactor fuel to facilitate its recycle and disposition. This process involves chemical and electrochemical methods to separate and recover highly enriched uranium from the used fuel into a purified high-assay low-enriched uranium metal form.

This research focused on fuel dissolution and separation of its aluminum matrix and cladding. To provide a rapid demonstration of this step, neodymium metal was used as a non-radiological surrogate for uranium metal. Thus, the specific objective of this study was to demonstrate and characterize the dissolution of neodymium metal into a molten halide salt system while simultaneously removing aluminum as a volatile halide. Accordingly, aluminum metal particulate and foil (representing aluminum matrix and cladding, respectively) along with neodymium metal particulate were blended at bench scale with either ammonium chloride or bromide (halogenating agents) and lithium chloride or bromide, respectively, and heated. Ammonium halides decomposed into ammonia and hydrogen halides, the latter of which reacted with aluminum and neodymium metal to form their respective halides. The relatively high vapor pressures of aluminum halides drove their gaseous separation from the neodymium halides, which fused with their respective lithium halides. The demonstration yielded excellent separation of the metals with 98.2% and 97.8% of the loaded aluminum separating as volatile chlorides and bromides from their respective molten salt systems, while no neodymium or lithium halides were detected in their respective aluminum halide streams. Thus, this work proved the principle that aluminum metal can be effectively separated from neodymium metal via halogenation and volatile specie collection.

TALENT PIPELINE:
Steven Herrmann, student at University of Idaho

PUBLICATION:
Herrmann, S. D., H. Zhao, M. Shi, M. M. Jones, and M. N. Patterson, "Halogenation of used aluminum matrix test reactor fuel – a bench-scale demonstration with surrogate materials," *Journal of Nuclear Science and Technology* 59(3) (2021) 395–406.



(a) (b) (c)

Simplified sectional view of the furnace and off-gas trap assembly for series of halogenation and waste-form runs (a). Waste-form blend before (b) and after (c) consolidation.

Sensitive Methods for Organoiodide Speciation

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

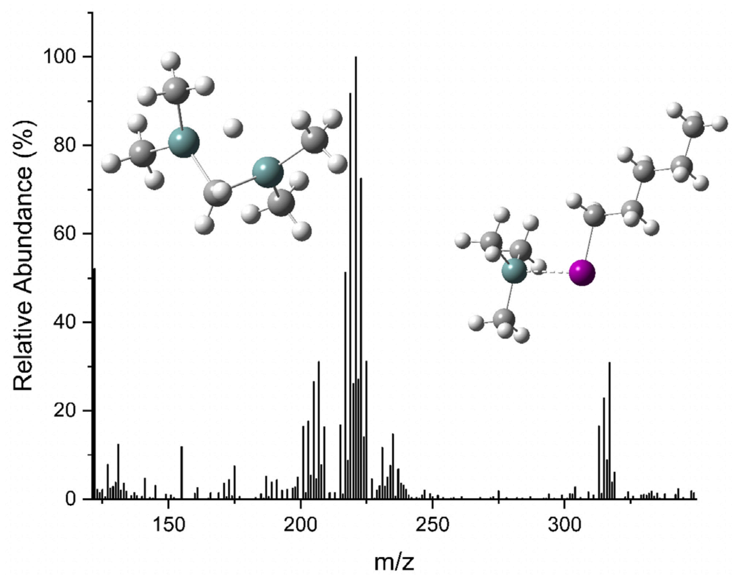
PROJECT NUMBER:
20A1049-008

PRINCIPAL INVESTIGATOR:
Christopher Zarzana

CO-INVESTIGATOR:
Gary Groenewold, INL

Novel research indicates that tetramethylgermanium could be used in the detection of organohalides in complex mixtures.

The purpose of this project was to investigate the utility of tetramethylgermanium as a chemical ionization reagent gas for the detection of organohalides using gas chromatography. Chemical ionization with tetramethylgermanium resulted in efficient formation of tetramethylgermanium -organohalide adduct ions. Additionally, initial evidence suggests that these adduct ions fragment less than the equivilent adducts formed when tetramethylsilane is used as the chemical ionization reagent gas, suggesting tetramethylgermanium may have utility in detection of organohalides in complex mixtures.



Mass spectrum of iodopentane ionized with chemical ionization using tetramethylgermanium as the reagent gas.

Accelerating Irradiated Fuel Studies by Advanced Surrogate Samples

TOTAL APPROVED AMOUNT:
$152,000 over 3 years

PROJECT NUMBER:
18A40-014

PRINCIPAL INVESTIGATOR:
Fidelma Di Lemma

CO-INVESTIGATORS:
Nathan Jerred, INL
Patrick Moo, INL
Robert O'Brien, INL
Tsvetoslav Pavlov, INL
Tiankai Yao, INL

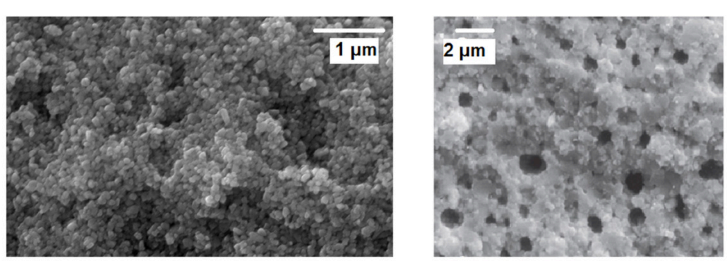
Advanced manufacturing techniques enable researchers to explore new approaches to accelerate and reduce the cost of nuclear fuel development.

Qualification of new nuclear fuel and the safe operation of conventional fuels require a deep understanding of fuel performance up to high burnup. Current fuel performance testing is mainly achieved by long and expensive irradiation campaigns, which are nearly impossible to accelerate in reactor. This research explored a new approach to accelerate and reduce the cost of fuel development through the fabrication of surrogate samples that mimic irradiated fuel. The realistic simulated irradiated fuel microstructure is possible by leveraging in-house fabrication capabilities, such as spark plasma sintering. These surrogates simulated both the high burnup structure and the chemical composition of irradiated fuel. These samples represent a unique opportunity to perform separate and combined effects studies, providing valuable validation data for the MARMOT microstructure evolution modeling suite.

The first proof-of-concept for these surrogates was obtained by producing uranium dioxide surrogate samples that simulated high burn up microstructure. Parameters of the powder preparation and spark plasma sintering method were optimized to reproduce grain size, reaching the desired 200 nm dimension. Porous formers were tested for the reproduction of the porosities observed in the high burnup structure. While the density, distribution, and volumetric concentration were in line with the ones found in irradiated fuel, the porosities seemed to be much smaller than the desired ones and required the use of much larger porous former. The final task was the introduction of surrogate fission products. This was successful as all the products were retained in the pellet. However, the distribution and size of the fission product precipitates was not in line with that reported in the literature. Further powder processing could be successful in producing smaller and better distributed precipitates. The reaction of fission products to form intermetallic and gray phase precipitates was also analyzed and was observed to be limited. Further annealing may be necessary to promote such reactions. Finally, thermal properties of these surrogates were measured to be compared to irradiated data.

TALENT PIPELINE:

- Patrick Moo, student at University of Florida
- Nathan Jerred, student at University of Idaho
- Tsvetoslav Pavlov, INL
  Russell L. Heath Distinguished Postdoc
- Tiankai Yao, Postdoc from Rensselaer Polytechnic Institute



(a) (b)

(a) Grain size obtained via spark plasma sintering, leading to desired grain structure. (b) Comparison with real irradiated high-burnup structure microstructure from Spino et al. (2012).

Coordinated Examination of Microstructure and Thermal Properties of Gamma Uranium-Zirconium Alloy in Volumetric Space

TOTAL APPROVED AMOUNT:
$124,862 over 1 year

PROJECT NUMBER:
20A1052-027

PRINCIPAL INVESTIGATOR:
Cynthia Adkins

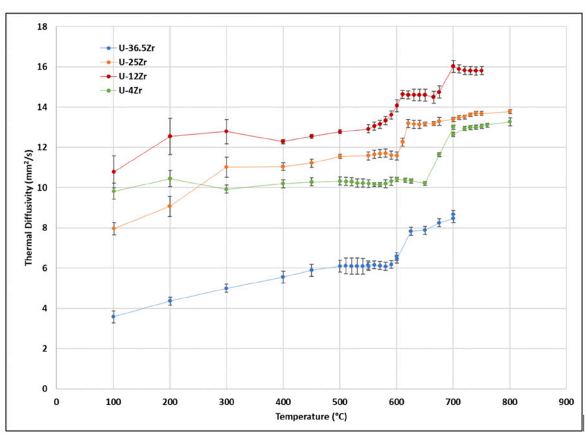
CO-INVESTIGATORS:
Andrea Jokisaari, INL
Tiankai Yao, INL
Zilong Hua, INL

Understanding the basic science principles governing thermal transport in metal fuel forms can enable new reactor designs.

The focus of this research was to characterize the gamma phase of uranium-zirconium (U-Zr) alloy for thermal conductivity and microstructure relationships. Material properties of the gamma phase are important to understand because the gamma phase is the dominant structure of the fuel during reactor operation. Thermal conductivity in metallic fuel systems during irradiation is affected by several factors, such as fission gas porosity formation, constituent redistribution and sodium infiltration into the periphery of the fuel. This investigation focused on the effect of constituent redistribution, mainly zirconium with temperature.

The gamma phase in this alloy system only exists above 600°C. Data was collected from room temperature to above 600°C to characterize the transition into the gamma phase. Bulk and local thermal conductivity were measured on the sample test matrix with some success. One of the challenges to collecting local thermal conductivity at high temperature on the U-Zr alloy was that as the sample approached the gamma phase transition temperature, the surface properties changed. This is likely due to the spinodal decomposition mechanism that occurs between the delta and gamma phases of U-Zr. This scattered the thermal conductivity microscope laser signal and made measurements very noisy and difficult to analyze. The spinodal decomposition mechanism was examined on similar U-Zr samples using the heated transmission electron microscope stage and the results of that study aided the thermal conductivity data analysis.

The thermal properties data collected on the gamma U-Zr system and its relationship to the microstructure are important because they inform the mechanistic understanding of the zirconium contributions in that alloy system. Therefore, these data are being incorporated into thermal transport models for irradiated metallic fuel by the Nuclear Energy Advanced Modeling and Simulation program.



Thermal diffusivity versus temperature for all alloy compositions in this study measured by laser flash analysis. Slope changes above 500°C indicate phase changes included on the U-Zr phase diagram.

Fast, High Resolution Autoradiographic Imaging of Localized, Small Fission Product Containing Samples for Nuclear Quality Assurance, Radiological Emergency Response, and Nuclear Forensics

TOTAL APPROVED AMOUNT:
$127,042 over 1 year

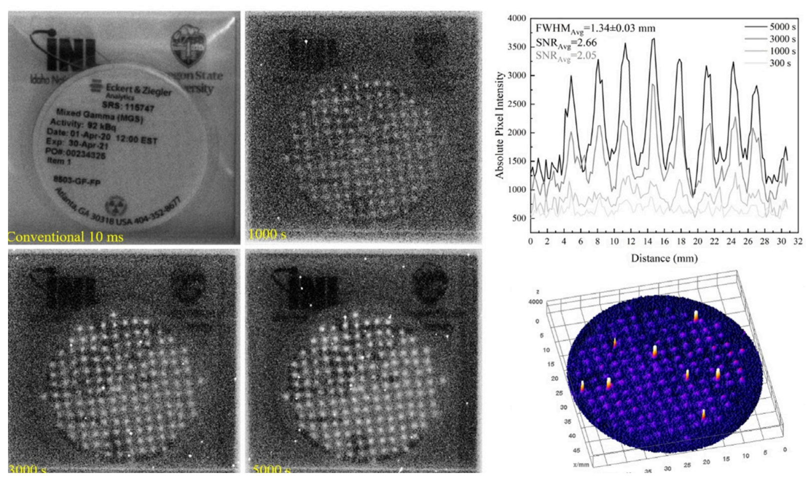
PROJECT NUMBER:
20A1049-016

PRINCIPAL INVESTIGATOR:
Ariana Foley

CO-INVESTIGATOR:
Julie Bowen, INL

Inexpensive and high-resolution autoradiographic imaging technology enables applications in nuclear quality assurance, radiological emergency response, and post-irradiation examination of materials.

This research developed and proved the principle of a novel autoradiographic method based on a bench-scale prototype integrating an electron-multiplying charged coupled device sensor and an inorganic scintillator-based method. Rapid analysis of the spatial distribution of radioactivity in samples demonstrated with the prototype will enable many applications in the nuclear industry, such as nuclear quality assurance, radiological emergency response, and post-irradiation examination of materials at a nuclear reactor or accelerator facility. This new capability addresses a limitation of current autoradiography methodologies in which image formation occurs directly from radiation emitted by the object itself, providing a two-dimensional image of the spatial distribution and relative intensity of radiation emitted from the analyzed sample. Traditional autoradiographic imaging methods of nuclear materials, such as film autoradiography or imaging plate technology, can be time-consuming and costly—problematic when samples must inform emergent safety decisions or capture essential time-dependent information.



(Left) A series of autoradiographic images of a 50 mm glass particulate filter compared to a conventional gain image at 1000, 3000, and 5000 s exposure times. The non-radioactive features such as the logos can be seen in the image. (Top right) A two-dimensional pixel intensity profile of the second from the top row with image quality assessments (i.e., full width half maximum and signal to noise ratio) of the autoradiographic images. (Bottom right) A three-dimensional intensity plot of a selection of the 5000 s exposure autoradiographic image that shows the collection of higher activity regions in the filter pores.

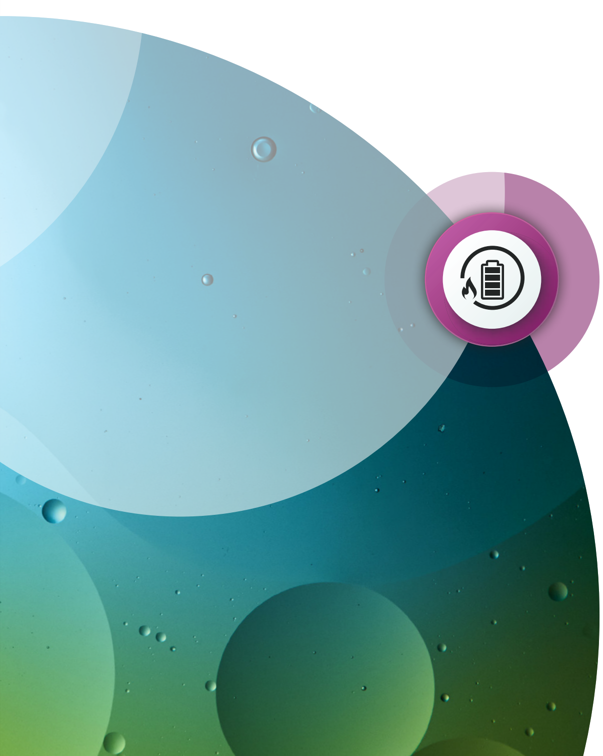
TALENT PIPELINE:

- Ariana Foley, student at Oregon State University

Proof-of-principle was achieved by completing a series of imaging experiments with known activity calibration sources and fission product doped spheroid surrogate nuclear debris particulate on the micrometer to millimeter scale. These experiments determined the optimal imaging parameters and experimental setup to provide high-quality autoradiographic images. Further experiments determined that both autoradiographic and non-radioactive features could be distinguished from a single, rapid exposure using this method. These results establish that the methodology has a high potential to be a novel contribution to autoradiographic imaging technology as an alternative to conventional methods that may require multiple exposures, additional instrumentation, and post-processing to compare non-radiographic with autoradiographic images of a single sample. The investigated novel autoradiographic imaging method has the potential to benefit a multitude of autoradiography applications by shortening the timing for traditional autoradiography analysis of radioactive samples from the current 12-78 hours timeframe to the timescale of seconds to minutes while providing spatial information on the non-radioactive features within the field of view.

INTEGRATED ENERGY SYSTEMS

INL leads RD&D to achieve a future in which energy demands across multiple energy use sectors are met by a combination of non-emitting energy sources (e.g., nuclear, renewable, fossil with carbon capture and sequestration/utilization). Innovative technology solutions that use nuclear energy products (e.g., heat, electricity, radiation) in coordination with other clean energy sources will accelerate the deployment of an economy based on affordable, clean, reliable, and sustainable energy. To realize sustainable and economically competitive net-zero integrated energy systems of the future, innovations are needed across generation, delivery, storage, and end use.



Nuclear energy is a near-zero air emissions option that can reliably meet electricity demands, but its value extends far beyond electricity generation: it is capable of dispatching both heat and electricity. INL RD&D focuses on developing multigeneration energy systems that incorporate nuclear heat with all forms of electricity generation to produce food, fuels, chemicals, construction materials, metals, and electronic devices, as well as power for heating and communications. Direct use of the radiation that is produced by nuclear energy is a new frontier for industry applications. Concepts for using radiation to catalyze chemical and materials changes remains at a conceptual stage. This effort will accelerate technology development and deployment across diverse geographical regions of the U.S., with many projects and technologies being demonstrated first at INL to advance towards a net-zero laboratory campus, while concurrently partnering with clean industrial complexes, and regional hydrogen and carbon management hubs that are part of the evolving electricity grid and energy infrastructure of the country.

High-performance Computing-based Dynamically Adaptive Protection Schemes for Electric Grid

TOTAL APPROVED AMOUNT:
$990,529 over 3 years

PROJECT NUMBER:
18A12-212

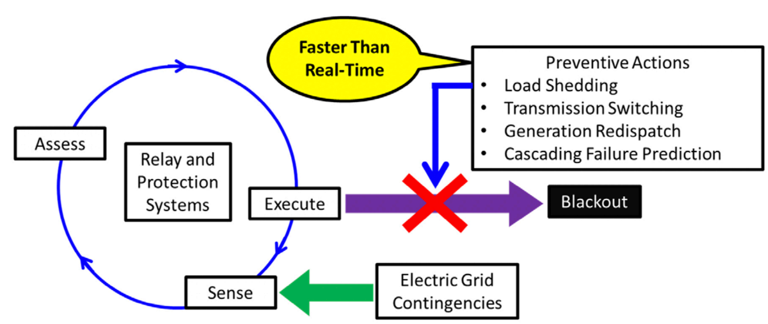
PRINCIPAL INVESTIGATOR:
S M Shafiul Alam

CO-INVESTIGATORS:
Siddharth Suryanarayanan, South Dakota State University
Svetlana Poroseva, University of New Mexico

COLLABORATOR:
Idaho Power Company

Faster than real time predictive capabilities lead to a secure and resilient electric grid.

The electric grid is becoming more complex, more dynamic, and less predictable due to the integration of newer technologies and approaches driven jointly by science and policy. These forces increase the risk of cascading failures and, therefore, compromise electric grid resiliency. Two major challenges of the future grid are: (1) the prediction and mitigation of cascading failures; and (2) the real-time adaptive methods for protection and remedial action schemes. A remedial action scheme is a procedure designed to activate under certain system stress conditions that may not exceed individual component protection thresholds. Practical solutions for dynamic reconfiguration require faster than real time predictive capabilities to analyze thousands of scenarios every few microseconds. In this project, the team developed a high-performance real-time co-simulation platform by integrating a digital real-time simulator and high-performance computing for outage prediction and dynamically adapting the protection settings. Optimal operation scenarios were identified, and the protection and remedial action schemes were dynamically optimized based on the real-time state of the electric grid. Research suggests a shift from state-of-the-art static to dynamic settings for wide area protection. An improved and less conservative protection and remedial action scheme operation was obtained by optimizing settings dynamically. Such methods incorporate more dynamic behavior in protection, control, and restoration and are applicable to advanced energy infrastructure.



The high-performance computing simulation platform enables the validation of any power system contingency (e.g., blackout) mitigation scheme, such as load shedding and transmission switching in real-time.

TALENT PIPELINE:

- Mohit Sinha, INL postdoc
- Bhaskar Mitra, INL postdoc
- Tanveer Hussain, student South Dakota State University
- Phillip Scott, student at University of New Mexico

PUBLICATION:

Hussain, T., S. M. Shafiul Alam, T. M. Hansen, and S. Suryanarayanan, "The LSBmax algorithm for boosting resilience of electric grids post (N-2) contingencies," *IET Journal of Engineering* 2021(12) (2021) 807–816.

PRESENTATIONS:

Sinha, M., R. G. Kadavil, M. Panwar, T. Hussain, S. Suryanarayanan, S. M. Shafiul Alam, and M. Papic, "An integrated high-performance computing and digital real-time simulation testbed to benchmark closed-loop load shedding algorithms in power systems," *The 52nd North American Power Symposium*, College Station, TX, USA, Apr. 11 - 14, 2021.

Hussain, T., S. M. Shafiul Alam, T. M. Hansen, and S. Suryanarayanan, "A computationally improved heuristic algorithm for transmission switching using line flow thresholds for load shed reduction," 14th *IEEE Madrid PowerTech Conference 2021*, Madrid, Spain, Jun. 28 - Jul. 2, 2021.

Poroseva, S. V., K. R. Soules, S. M. Shafiul Alam, M. Panwar, and R. Hovsapian, "A benchmark case for the grid survivability analysis," *IEEE PES General Meeting 2021*, Virtual, Jul. 26–29, 2021.

Hussain, T., S. Suryanarayanan, and S. M. Shafiul Alam, "An improved transmission switching algorithm for managing post-(N-1) contingencies in electricity networks," *International Conference on Applied Energy (MIT A+B)*, Massachusetts Institute of Technology, Cambridge, MA, USA, Aug. 13-14, 2020.

INTELLECTUAL PROPERTY:

Hussain, T., S. Suryanarayanan, and S. M. Shafiul Alam, "Hybridized transmission switching for contingency management in electric power systems," International Patent Application: PCT/US21/26540, patent pending (April 9, 2021).

Fundamental Liquid/Porous-Solid Interactions in High-solids Slurry Processing

TOTAL APPROVED AMOUNT:
$707,500 over 2 years

PROJECT NUMBER:
19A39-185

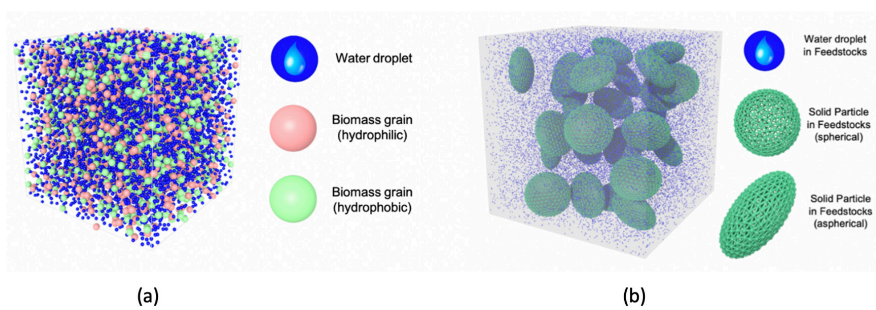
PRINCIPAL INVESTIGATOR:
Luke Williams

CO-INVESTIGATORS:
Yidong Xia, INL
Jiaoyan Li, State University of New York at Buffalo

Understanding the rheological and interfacial properties of high-solids slurries as a function of their physical and chemical characteristics reduces manufacturing waste and energy demand.

Processing high-solids slurries, wet solids slurries containing >50% solids by volume, is a challenge for many industries, such as converting biomass to fuels or products, food processing, electrode manufacturing for batteries, and the formation of photovoltaics and other sensors. These processing challenges often arise from a lack of fundamental understanding of the coupled particle-particle mechanical and particle-fluid interfacial interactions that dominate the non-Newtonian rheology of these systems. Understanding the rheological and interfacial properties of these slurries as a function of the physical and chemical characteristics of the system would allow manufacturers to achieve more consistent products with reduced waste and energy demands.

A primary reason for the processing difficulty of wet solids stems from significant variability in the solids fraction of the slurry in terms of surface chemistry, particle size, size distribution, and morphology, which all play a major role in determining both solid-solid and solid-liquid interfacial interactions. This project addressed these challenges by characterizing the solids physical properties and free chemicals in the slurry system that affect the solid-liquid interactions. These impactful physical and chemical properties were also manipulated by using treatments during material storage and preprocessing to alter the properties of interest. System characteristics are then correlated to viscosity data measured through capillary rheology experiments that mimicked process relevant conditions. Additionally, the material characterization and processing experiments were used as baseline data to inform the development and validation of a computational model for slurry.



Dissipative particle dynamics configurations of high-solids slurries systems: (a) Solid particles without self-structure described explicitly, in which the blue particles represent water droplets (with radius of 20μm), pink particles represent biomass grains (with radius of 50 μm) with hydrophilic surface properties and green particles represent biomass grains (with radius of 50 μm) with hydrophobic surface properties. (b) Solid particles with self-structure described explicitly, in which the blue particles represent water droplets, and green particles of spherical and aspherical shapes.

TALENT PIPELINE:

- Carl Fredrik Mikael Karlsson, INL postdoc

The slurry model is based on a novel mesoscale dissipative particle dynamics method that accounts for both the physical and chemical characteristics of a slurry system through the particle-particle and particle-fluid interactions. The dissipative particle dynamics model can be used to probe how the changes in the particle-fluid interactions alter slurry viscosity of a complex non-Newtonian fluid system.

This research used capillary rheology experiments to show how different treatments lower viscosity by over 150x or increase it by 2x. Torrefaction made pine particles so friable that the small final particle size and hydrophobicity cause the sample to flow much more smoothly in the capillary system. Dimethyl ether pretreatment of pine to remove extractives increased the sample viscosity by a factor of two compared to the control sample despite having statistically identical particle sizes. Biological degradation of the corn stover decreased viscosity by about a factor of three compared to the control stover, which bodes well for improving the viscosity of industrial samples like the solids left over after anaerobic digestion.

Intensified Characterization and Analysis of Energy Storage System to Support Integrated Energy Systems

TOTAL APPROVED AMOUNT:
$1,894,656 over 3 years

PROJECT NUMBER:
19P45-013

PRINCIPAL INVESTIGATOR:
Boryann Liaw

CO-INVESTIGATORS:
Bor-Rong Chen, INL
Gorakh Pawar, INL
John Koudelka, INL
Meng Li, INL
Qiang Wang, INL
Ross Kunz, INL
Leslie Kerby, Idaho State University

Machine learning-based analytics to identify and quantify failure modes and effects enable intelligent approach to battery energy storage systems development.

Battery energy storage systems play a pivotal role in the advancements of the portable electronics and Internet of Things, the electrification of transportation and mobility, and the energy efficiency and security in the supply chain of the power grid. To enable more reliable functionalities of lithium-ion battery energy storage systems to support this intensified electrification in the energy supply chain, basic understanding of how to improve the durability, performance, reliability, and safety of the lithium-ion battery energy storage systems is vital. Development of a consistent platform to evaluate, diagnose, and assure the quality of the battery and its function through the entire life cycle—from material preparation, component and battery design, fabrication, and control and management of the lithium-ion battery energy storage systems in operation is needed to yield better functionalities.

This project developed a quantitative failure mode and effect analysis platform that can enhance battery diagnostic and prognostic capability with advanced data analytic and regression techniques. The approach combined quantitative failure analyses and separated algorithmic and mechanistic model regressions and predictions, respectively, using advanced artificial intelligence and machine learning-based analytics to identify failure mechanisms, quantify the effects, verify the analyses, and validate the predictions. This novel approach achieved a high precision and accurate analytic platform for future high-throughput data handling and high-performance computing-based failure mode and effect analysis processes.

TALENT PIPELINE:

- Yulun Zhang, INL postdoc
- Yuxiao Lin INL postdoc
- Jim Greene, student at University of Idaho
- Steven McDaniel, student at Idaho State University
- Shovan Chowdhury, student at Idaho State University

INTELLECTUAL PROPERTY:

Liaw, B., Y. Zhang, and Q. Wang, "Energy storage cell qualification and related systems, methods, and devices," U.S. Patent Application 17/149,046 (January 14, 2021).

Liaw, B., Y. Zhang, and Q. Wang, "Electrochemical analytic diagnosis for battery cell qualification," U.S. Patent Application 62/961,096 (January 14, 2020).

AWARDS:

- 2020 Technology Award, International Battery Materials Association
- 2019 Asian American Engineer of the Year

PUBLICATIONS:

Zhang, Y., R. Nguyen, and B. Liaw, "Status and gaps in rechargeable lithium battery supply chain: Importance of quantitative failure analysis," Proceedings of the *IEEE* 109(6) (2021) 1029–1038.

Zhang, Y., Q. Wang, B. Liaw, S. C. Nagpure, E. J. Dufek, and C. C. Dickerson, "Cell qualification—A performance metric-based approach," *Journal of Physics: Energy* 2(3) (2020) 034003.

Zhang, Y., Q. Wang, B. Liaw, S. C. Nagpure, E. J. Dufek, and C. C. Dickerson, "A quantitative failure analysis on capacity fade in rechargeable lithium metal cells," *Journal of the Electrochemical Society* 167 (2020) 090502.

Mattei, G. S., Z. Li, A. A. Corrao, C. Niu, Y. Zhang, B. Liaw, C. C. Dickerson, J. Xiao, E. J. Dufek, and P. G. Khalifah, "High-energy lateral mapping (HELM) studies of inhomogeneity and failure mechanisms in NMC622/Li pouch cells," *Chemistry of Materials* 33(7) (2021) 2378–2386.

Wang, F., Z. Lin, L. Liu, X. Wei, S. Lin, L. Dai, Y. Wei, C. Liang, and B. Liaw, "Does polarization increase lead to capacity fade?," *Journal of the Electrochemical Society* 167 (2020) 090549.

PRESENTATIONS:

Li, M., C. Fang, Y. Zhang, B. Lu, Y. S. Meng, G. Pawar, Y. Lin, and B. Liaw, "Can fast charging rechargeable lithium batteries be a reality?" *Symposium A02 – LIBs Fast Charging, 239th Electrochemical Society Meeting*, Virtual, May 30–Jun. 3, 2021.

Zhang, Y., B. Liaw, E. J. Dufek, and B. Li, "First cycle capacity loss of Li–NMC 811 Cells—A close look from electrochemical diagnostic analysis," A special seminar presented to personnel at the GM Warren Technical Center, Warren, MI, USA, Mar. 2, 2021.

Zhang, Y., B. Liaw, E. J. Dufek, and B. Li, "Tracking rechargeable Li battery cell degradation through quantifiable capacity fade modes from formation to cycle aging," *2021 IAPG Chemical Working Group (CWG) Safety Panel Virtual Meetings*, Virtual, Feb. 19, 2021.

B. Liaw, et al., "Quantitative cell qualification to enable long life rechargeable Li metal batteries." *International Battery Association 2020* (IBA-2020), Nov. 17–18, 2020.

B. Liaw, "INL's battery diagnostic and prognostic approach for failure mode and effect analysis (FMEA)—Identify cell quality through better qualification," *Battery Safety Council Forum 9*, Nov. 17–18, 2020.

Zhang, Y., B. Liaw, E. J. Dufek, and B. Li, "How well cathode materials are being used in rechargeable Li batteries?" *PRiME 2020, 238th Electrochemical Society Meeting, Symposium of Intercalation Chemistry for Electrochemical Energy Storage Technologies*, Virtual, Oct. 4–9, 2020.

Liaw, B., “Lithium metal electrode—Stability, reversibility, and durability,” *Li Metal Electrode Characterization and Modeling Workshop*, University of California–San Diego, La Jolla, CA, USA, Feb. 13, 2020.

Liaw, B., “From battery failure analysis to life prediction,” *Battery Innovation Summit, NAATBatt Annual Meeting and Conference 2020*, Pasadena, CA, USA, Feb. 12, 2020.

Liaw, B., “Quantify rechargeable Li metal cell performance through failure analysis and multi-scale modeling,” *Department of NanoEngineering Seminar*, University of California–San Diego, La Jolla, CA, USA, Nov. 27, 2019.

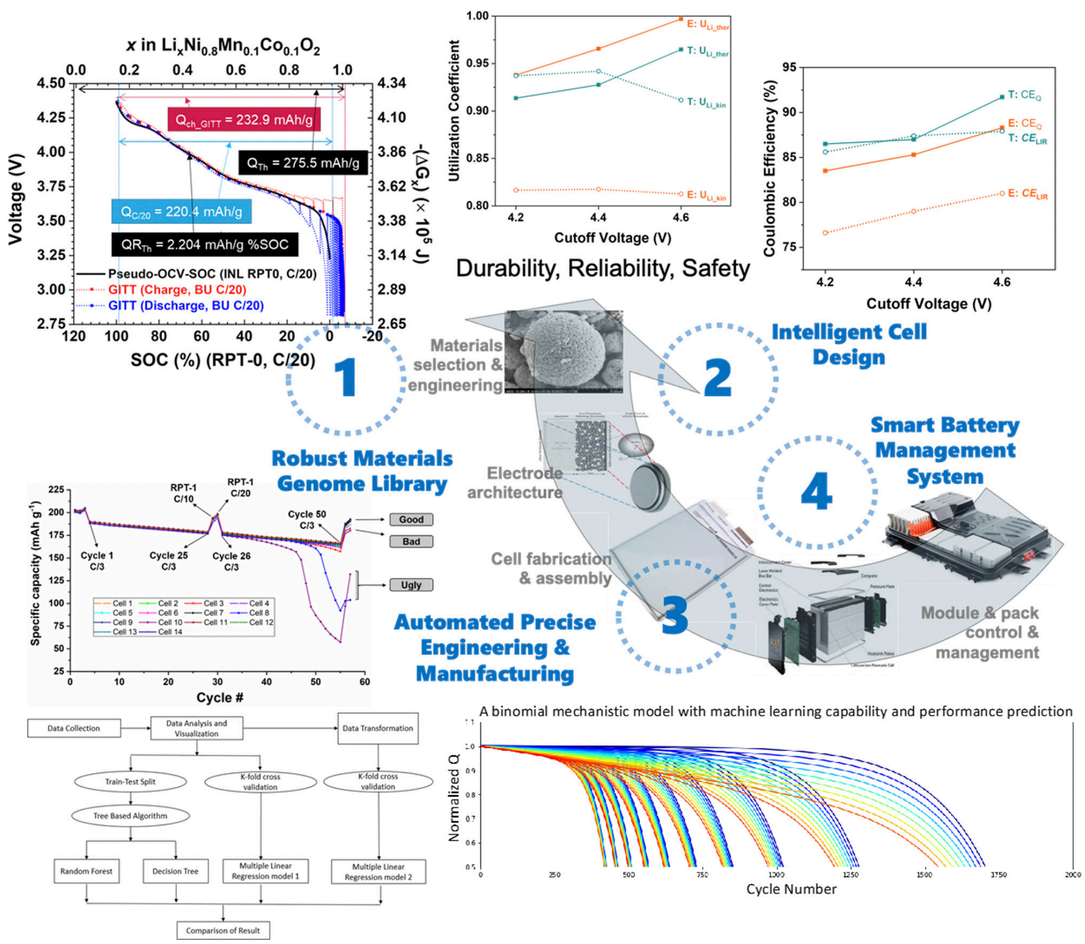
Zhang, Y., and B. Liaw, “From battery failure analysis to safety mitigation.” *Battery Safety Council Forum #8*, Washington, D.C., USA, Nov. 19–20, 2019.

Liaw, B., “Lithium metal electrode — Understanding its characteristics,” *Department of Chemistry Seminar*, Temple University, Philadelphia, PA, USA, Nov. 11, 2019.

Liaw, B., Y. Zhang, Q. Wang, E.J. Dufek, B. Li, G. Pawar, N. Gao, and S. Kim, “Failure mode and effect analysis for rechargeable lithium battery safety,” *The Third International Battery Safety Workshop* (IBSW-2019), Beijing, China, Oct. 8, 2019.

Liaw, B., “Battery technology and safety,” *2019 CIE/USA-DFW and AAEOY International Technology & Leadership Conference (ITLC)*, Dallas, TX, USA, Aug. 17, 2019.

Liaw, B., “Can failure mode and effect analysis redefine battery safety?” *GM Warren Technical Center*, Warren, MI, USA, Mar. 26, 2019.



Intelligent analysis-qualification-design-verification loop developed to achieve quantitative failure mode and effect analysis and establish a smart diagnostic-prognostic platform for automated battery manufacturing capability in the future.

Innovative Materials and Coatings for Sweep Gas Membrane Distillation to Separate Bioethanol from Fermentation Broth

TOTAL APPROVED AMOUNT:
$115,000 over 1 year

PROJECT NUMBER:
20A1047-018

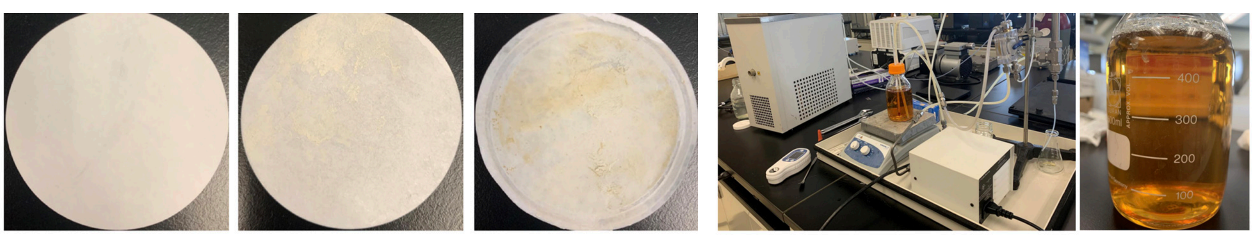
PRINCIPAL INVESTIGATOR:
Birendra Adhikari

CO-INVESTIGATORS:
Aaron Wilson, INL
Christopher Orme, INL
John Klaehn, INL

Novel membrane chemistry and separations process can reduce the cost of the energy required to produce ethanol by 30% as compared to traditional distillation separation.

The objective of this project was to develop an innovative, robust, and economically viable omniphobic (repels all compounds) membrane for a sweep gas membrane distillation process that uses low grade heat sources (such as nuclear or solar heat) to recover/separate bioethanol from the slurry after fermentation. The membrane is part of a fouling resistant ethanol separation process that can reduce the cost of bioethanol separation by producing game-changing improvements to membrane distillation through advanced membrane materials. Researchers hypothesized that a novel molecular surface layer of an omniphobic compound could be attached to the membrane surface in a way that maintains the mechanical properties and porosity of the membrane and improve the membrane performance (permeance and fouling resistance).

An omniphobic membrane was developed that is porous and has fouling resistant properties, creating a more efficient and cost-effective membrane for bioethanol-water separation. This membrane was prepared by the layer-by-layer deep coating of a polyvinylidene difluoride membrane material and 1H,1H,2H,2H-perfluorodecyltriethoxysilane as an omniphobic surface coating. A commercially available membrane was purchased, tested, and characterized as the starting membrane material. Forward osmosis and membrane distillation membranes were built in-house as part of this project and were used for the separation tests. In-house nitrogen gas was used as the sweep gas for these experiments. Concentrated aqueous slurries containing: (1) 10% by weight of ethanol only; and (2) fermentation broth with 10% by weight ethanol were used as feeds for the membrane distillation process. The clean and concentrated ethanol was recovered as the product. It was found that the commercially available membrane had ethanol-water selectivity of 2.2 to 2.5 and the membrane developed under this research has ethanol-water selectivity of 3.2 to 3.5. A technoeconomic analysis model developed as a part of this project shows that these omniphobic membranes can reduce the cost of the energy required to produce ethanol by 30% as compared to traditional distillation separation.



(a) (b) (c) (d) (e)

(a) Commercially available membrane, (b) modified ominphobic membrane, (c) omniphobic membrane after use, (d) membrane system, and (e) fermentation broth.

Microalgae for Selective Concentration of Rare Earth Elements from Bioleachate Solutions

TOTAL APPROVED AMOUNT:
$101,535 over 1 year

PROJECT NUMBER:
20A1047-025

PRINCIPAL INVESTIGATOR:
Bradley Wahlen

CO-INVESTIGATOR:
David Reed, INL

COLLABORATORS:
Pacific Northwest National Laboratory
Arizona State University
Washington State University

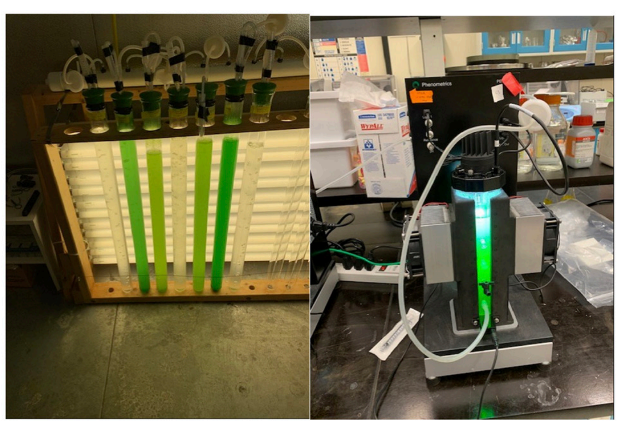
An environmentally friendly process to extract rare earth elements from secondary sources could advance efforts to establish a sustainable domestic supply of these elements.

Rare earth elements are essential to implementing clean energy technologies, and their supply is at risk due to increased demand, the limited number of commercially accessible natural deposits, and a fragile supply chain. These challenges have generated interest for recovering rare earth elements from secondary sources, such as electronic waste and industrial catalysts. INL has developed a bioleaching approach for rare earth element recovery that is economically viable and environmentally sustainable. Like traditional acid dissolution, bioleaching solubilizes various metal ions in addition to rare earth elements, requiring effective down-stream separations for successful recovery.

This research explored using acidophilic microalgae to selectively remove rare earth elements from solution through sorption-mediated processes when cultivated in bioleachate solutions resulting from the processing of electronic waste. Nine acidophilic strains of microalgae were obtained from culture collections and collaborators. They were cultivated in a common media to facilitate experimentation. Of the nine strains, two did not acclimate to the media and were lost. The remaining seven strains were used in experiments to determine whether mixotrophic growth on gluconic acid is possible and whether they could facilitate biosorption of targeted elements when cultivated in media supplemented with metals to simulate a leachate obtained from recycling neodymium magnets. Gluconic acid is a key constituent of a biolixiviant produced by Gluconobacter oxidans that effectively solubilizes metals from electronic waste. It was found that gluconic acid severely reduce algae growth rates, at lower concentrations. Therefore, a longer duration research project that has the time to optimize cultivation conditions will be needed to thoroughly evaluate the concept of recovering rare earth elements through acidophilic microalgae biosorption.

TALENT PIPELINE:

- Caitlin McNamara, student at Idaho State University
- David Gazzo, student at Montana State University



Environmental photo bioreactor used for biosorption studies.

Solid State Chlorination of Waste Lithium Battery Scrap

TOTAL APPROVED AMOUNT:
$108,000 over 1 year

PROJECT NUMBER:
20A1049-029

PRINCIPAL INVESTIGATOR:
Mark Strauss

CO-INVESTIGATORS:
Luis Diaz Aldana, INL
Mary Case, INL
Tedd Lister, INL

Novel chemistry expands critical material recovery and supports a more sustainable decarbonized energy grid.

This research developed an innovative pathway for high purity extraction of cobalt, lithium, nickel, and manganese from end-of-life lithium-ion batteries. Solid-state chlorination was used to: (1) remove impurities; (2) use fewer and less toxic reagents; and (3) reduce the number of unit operations in lithium-ion battery scrap leaching and purification. Solid-state chlorination effectively extracts cobalt and lithium from permanent magnets and cathode material of lithium-ion batteries. The intention of this research project was to demonstrate that the technology can be extended to end-of-life batteries where impurities such as aluminum and iron can be removed and recovered separately. Ammonium chloride with low temperature roasting created a water-soluble metal chloride concentrate ready for metal separation. After assembly of the experimental equipment, Box-Behnken experimental design with three variables and three levels was performed to study the effect of temperature, gas carrier flow rate, and ammonium chloride to metal extraction and impurity removal efficiencies. The results show that over 92% extraction of cobalt, lithium, nickel, and manganese can be achieved in just 20 minutes.

TALENT PIPELINE:

- Mark Strauss, INL postdoc

PRESENTATION:

Strauss, M., L. D. Aldana, M. Case, and T. Lister, "A strategy for acid-free waste lithium battery processing," *Ni-Co 2021: The 5th International Symposium on Nickel and Cobalt*, The Minerals, Metals & Materials Society, Virtual, Mar. 15–18, 2021.

Metal	Weight %
Cobalt	14.16
Nickel	4.27
Manganese	3.31
Lithium	2.31
Aluminum	0.48
Iron	0.16
Copper	0.16
Zinc	0.09

The starting material from lithium-ion batteries for metal extraction and separation accounts for >50% of the battery weight. The table shows the weight percent of each metal in the starting material.

Macromolecule Radiolysis Using Radical Precursors

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PROJECT NUMBER:
20A1049-007

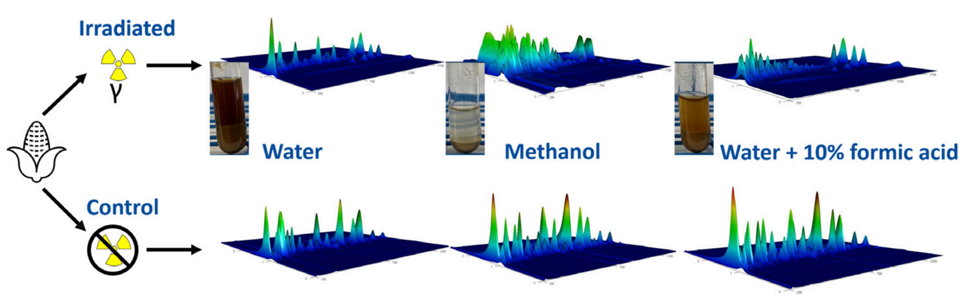
PRINCIPAL INVESTIGATOR:
Brittany Hodges

CO-INVESTIGATORS:
Christopher Zarzana, INL
Gary Groenewold, INL

Harnessing ionizing radiation to convert energy to molecules adds value to low-value macromolecules as they are broken down into smaller, more easily separated feedstocks or fuels before being converted to a value-added product.

The net-zero energy future depends on reusing carbon-based macromolecules and recycling waste products into higher value products. Converting macromolecules into liquid fuel or chemical feedstocks is challenging due to their large and sometimes heterogeneous network of polymeric bonds. This project tested the hypothesis that macromolecular materials can be infused with radical capping donors, and gamma irradiation of the resulting mixtures will produce ensembles of lower-molecular weight chemicals that can be more easily separated. The conversion chemistry was optimized by adjusting feedstock composition, irradiation time, and choice of radical capping donor to explore the different impacts of each of these materials on the radiolysis products. The results show that radiolysis of macromolecules in the presence of radical capping donors can break down complex macromolecules into smaller molecules while controlling re-polymerization. This research offers a new cost-efficient way to use radiation and heat byproducts of nuclear energy production to convert a simple mixture of complex molecules into a complex mixture of small molecules that can be used as an energy source or as potential feedstocks for new polymers or commodity chemicals.

This project demonstrated foundational molecular research for a potential new application of waste radiation and heat energy from nuclear energy production to convert waste products like plastics and biomass into potential new feedstocks to help offset the cost of spent fuel storage and management. This project offers a new process for converting lower value materials that are not cost-competitive or come with undesirable negative environmental impacts into higher-value feedstocks for commodity chemicals or liquid energy sources.



Control and irradiated sample comparison. Solvents tested were water, methanol, and water + 10% formic acid. Comprehensive gas chromatography mass spectroscopy shows significant differences between irradiated and unirradiated samples, with the most spectral changes in the methanol sample.

TALENT PIPELINE:

- Grace A. Castle, student at University of Notre Dame
- Samuel Tyndall, student at Northwestern University

PRESENTATIONS:

Hodges, B., C. Zarzana, G. Horne, and G. Castle, "Macromolecule gamma-radiolysis for polymer upcycling," *American Society for Mass Spectrometry (ASMS) Annual Conference*, Philadelphia, PA, USA, Oct. 31–Nov. 4, 2021.

Castle, G., B. Hodges, C. Zarzana, G. Groenewold, and G. Horne, "Pyrolysis-two-dimensional gas chromatography-mass spectrometry for characterization of irradiated biomass," *American Society for Mass Spectrometry (ASMS) Annual Conference*, Philadelphia, PA, USA, Oct. 31–Nov. 4, 2021.

Castle, G., and B. Hodges, "Co-radiolysis of corn stover biomass and solvent," *American Society for Mass Spectrometry (ASMS) Annual Conference*, Philadelphia, PA, USA, Oct. 31–Nov 4, 2021.

Switchable Solvent Water Extraction and Water Softening

TOTAL APPROVED AMOUNT:
$1,250,000 over 2 years

PROJECT NUMBER:
20A44-040

PRINCIPAL INVESTIGATOR:
Aaron Wilson

CO-INVESTIGATORS:
Christopher Orme, INL
John Lienhard, Massachusetts Institute of Technology

COLLABORATOR:
Massachusetts Institute of Technology

Discoveries fill knowledge gaps on fundamental thermodynamics important for process efficiency of transformative water treatment technologies.

Solvent-driven water-selective extraction zero liquid discharge desalination and solvent-driven fractional precipitation scalant removal are two interrelated water treatment technologies, each with the potential to transform multiple industries via reduced cost water treatment and recycling. Both technologies are projected to be energy efficient treatments that can address complex solutions containing multiple contaminants over a wide range of pH and concentrations. Both technologies are expected to isolate currently inaccessible products from waste streams, including: (1) mine drainage; (2) landfill leachate; (3) various scrubber blow downs, including flue gas desulfurization; (4) refining process waters (e.g., gas to liquid and coal to chemical); (5) produced waters from oil and gas production; (6) down-stream chemical production waters; (7) membrane system rejectates (including industrial manufacturing waters); (8) food processing; (9) mineral processing waters; and (10) pretreatment of existing desalination processes.

This project focused on fundamental thermodynamics that determine the process efficiency of the technologies. Filling these technical knowledge gaps is needed to develop a general technoeconomic model and understand commercial viability of the process. Additionally, this work produced data associated with solution and electrolyte theory to advance the current state of knowledge.

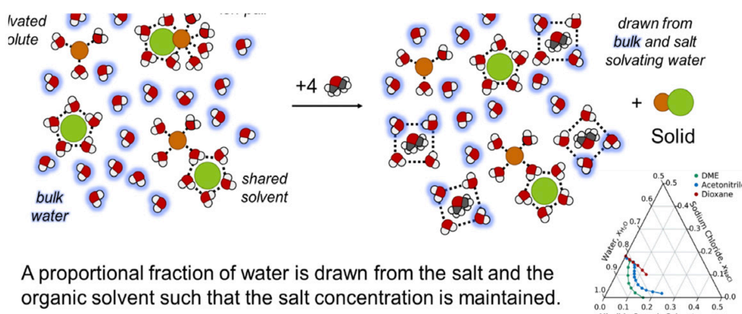


Illustration of the mechanism by which solvent-driven fractional precipitation occurs based on the mass-action solution model developed in this project, starting form a saturated sodium chloride solution. (inset bottom right) Select phase boundary data for water sodium chloride organic mixtures that aligns with the model.

TALENT PIPELINE:

- Zi Hao Foo, student at Massachusetts Institute of Technology
- Akshay Deshmukh, postdoc at Massachusetts Institute of Technology
- Caleb Stetson, INL postdoc
- Hyeonseok Lee, INL postdoc

PUBLICATIONS:

McNally, J. S., Z. H. Foo, A. Deshmukh, C. J. Orme, J. H. Lienhard, and A. D. Wilson, "Solute displacement in the aqueous phase of water–NaCl–organic ternary mixtures relevant to solvent-driven water treatment," *RSC Advances* 10 (2020) 29516–29527.

Wilson, A. D., C. Stetson, "Modeling solution vapor equilibria with solvation and solute assembly," *Journal of Molecular Liquids* 336 (2021) 116272.

Feeley, B. P., M. A. Overton, M. M. Galloway, T. J. Lecrivain, and A. D. Wilson, "Idaho database of solution thermodynamics," *Journal of Molecular Liquids* 338 (2021) 116574.

Wilson, A. D., H. Lee, and C. Stetson, "Mass action model of solution activity via speciation by solvation and ion pairing equilibria," *Communications Chemistry* 4 (2021) 163.

Deshmukh, A., Z. H. Foo, C. C. Stetson, H. Lee, C. J. Orme, A. D. Wilson, and J. H. Lienhard, "Thermodynamics of solvent-driven water extraction from hypersaline brines using dimethyl ether," *Chemical Engineering Journal* 434 (2022), 134391.

PRESENTATIONS:

Wilson, A. D., "Thermodynamic energy requirements of CO_2 switchable solvents in osmotically driven membrane processes," *Pacifichem 2021*, Honolulu, HI, USA, Dec. 15–20, 2021.

Wilson, A. D., "Phase-boundary relationships essential to solvent based water treatments," *Pacifichem 2021*, Honolulu, HI, USA, Dec. 15–20, 2021.

Wilson, A. D., "Solvent-driven aqueous separations," *National Laboratories Monthly Energy-Water Exchange Meeting*, Virtual, Aug. 16, 2021.

Wilson, A. D., "A speciation-based solution model," *Invited talk for the National Alliance for Water Innovation Modeling and Simulations Topic Area Monthly Webinar Series*, Virtual, May 3, 2021.

Wilson, A. D., "Solvent based water treatment and separation," *National Alliance for Water Innovation Annual Meeting, Virtual*, Nov. 18–19, 2020.

Wilson, A. D., and D. Ginosar, "*Switchable solvent dewatering for recovery of critical materials," CMI Annual Meeting*, Virtual, Sept. 15, 2020.

INTELLECTUAL PROPERTY:

Lienhard, J. H., A. Deshmukh, Z. H. Foo, and A. D. Wilson, "Switchable system for high-salinity brine desalination and fractional precipitation," U.S. Patent Application 17/567,809 (Jan. 3, 2022).

The Idaho Database of Solution Thermodynamics is available to scientists worldwide at https://idst.inl.gov/Home/Introduction.

A New Approach to Cathode Interface Structure/Activity with Transient Kinetics

TOTAL APPROVED AMOUNT:
$539,000 over 2 years

PROJECT NUMBER:
20A44-050

PRINCIPAL INVESTIGATOR:
Zongtang Fang

CO-INVESTIGATORS:
Qiang Wang, INL
Rebecca Fushimi, INL

COLLABORATOR:
University of Alabama

New understanding of the fundamental chemistry of the next-generation high-energy lithium-ion batteries advances energy storage technologies through transient kinetic experiments.

High nickel content lithium-nickel-manganese-cobalt oxides ($LiNi_xMn_yCo_zO_2$, NMC) are promising candidates as cathode materials for next-generation high-energy lithium-ion batteries. Significant challenges associated with the degradation of nickel rich NMC cathode materials include gradual capacity loss, as well as safety concerns resulting from reduced thermal stability, which could lead to combustion at a high state-of-charge. The degradation of NMC cathode materials primarily comes from side reactions, cation mixing, transition metal dissolution, and gas evolution, such as carbon dioxide (CO_2) and oxygen. It has been reported that surface impurities of lithium carbonate are notably involved in parasitic surface reactions, as well as in CO_2 production. A key challenge in the development of robust and durable materials is a fundamental understanding of how structure and composition impact the surface reactivity.

Temporal analysis of products (TAP) and infrared spectroscopy were combined with density functional theory modeling to study the formation of surface carbonates and hydroxides via CO_2 and water adsorption on various NMC cathode materials. TAP is a pulse response approach for mechanistic investigation and precise kinetic characterization in heterogeneous catalysis. In this project, TAP was applied for the first time in the application area of battery materials. Various NMC material with different nickel ratios (x:y:z = 8:1:1, 6:2:2, 5:3:2, 4:3:3, 1:1:1), as well as coated NMC materials, were studied to determine the role of different metals on carbonate and hydroxide formation and the role of water on carbonate formation.

The reactivity of the material was found to increase with the nickel content while the capacity for CO_2 had a distinct composition dependence. It was found that water creates more active sites for CO_2 adsorption through hydrogen bonding of surface hydroxyls. Furthermore, water can stabilize the reactive surface and hydroxyl groups greatly decelerate the carbonate formation process. Detailed kinetics of these processes were reported. This fundamental kinetic characterization points to strategies for controlling the surface reactivity and stability of NMCs through exposure of select crystalline planes and precise control of water. This new understanding will be used in the future to enable superior design of the next-generation cathode materials.

TALENT PIPELINE:

- Caitlin Hanson, student at University of Alabama
- Matthew Confer, student at University of Alabama

PUBLICATION:

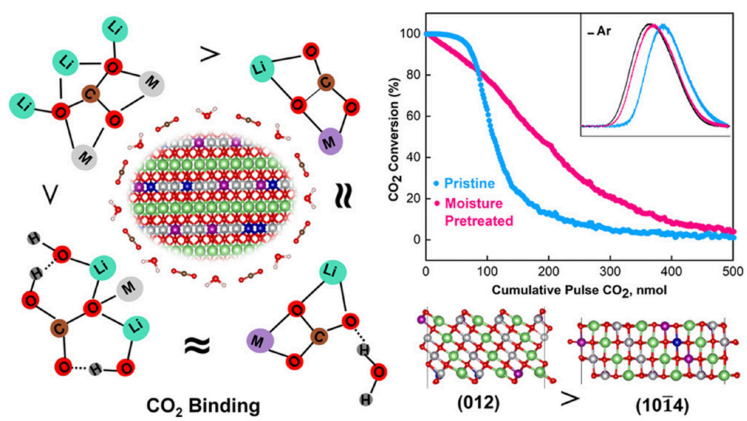
Fang, Z., M. P. Confer, Y. Wang, Q. Wang, M. R. Kunz, E. J. Dufek, B. Liaw, T. M. Klein, D. A. Dixon, and R. Fushimi, "Formation of surface impurities on lithium-nickel-manganese-cobalt oxides in the presence of CO_2 and H_2O," *Journal of the American Chemical Society* 143(27) (2021) 10261–10274.

PRESENTATIONS:

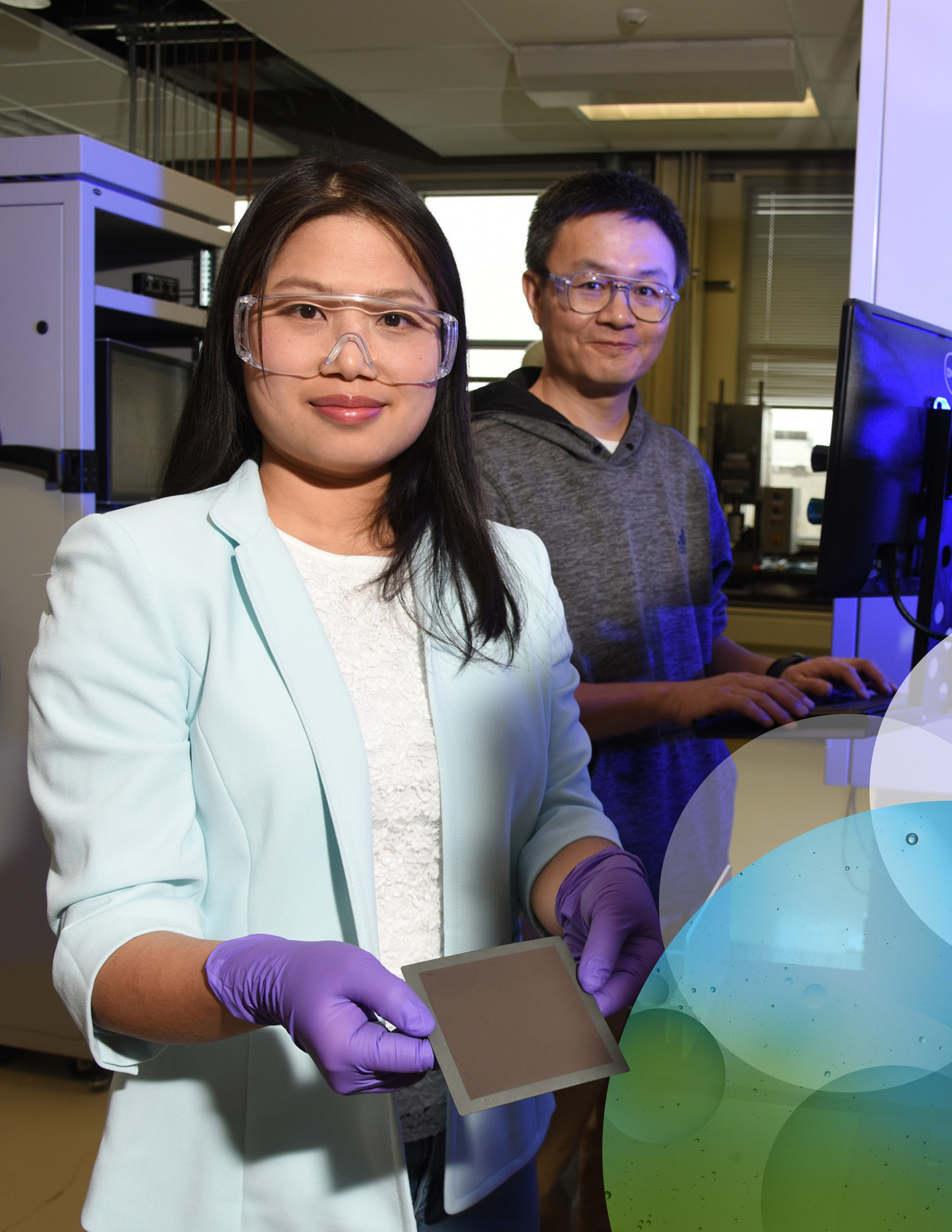
Fang, Z., et al., "Formation of surface impurities on Ni-rich NMC cathode materials due to CO_2 and H_2O," *International Chemical Congress of Pacific Basin Societies 2021*, Virtual, Dec. 16–21, 2021.

Fang, Z., et al., "Formation of surface impurities on Ni-Rich NMC cathode materials due to CO_2 and H_2O," *Transient Kinetics Seminar 2021*, Virtual, Apr. 26–28, 2021.

Fang, Z., et al., "Insight into the carbonate formation of Ni-Rich NMC cathode materials with CO_2," *AIChE Annual Meeting 2020*, Virtual, Nov. 16–20, 2020.

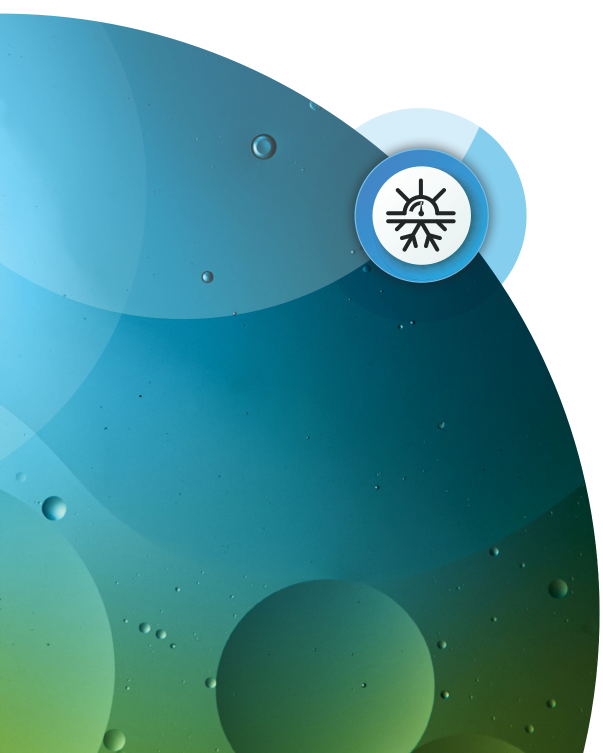


Surface impurities involving parasitic reactions and gas evolution contribute to the degradation of high nickel content NMC cathode materials. The TAP transient kinetic technique, density functional theory, and infrared spectroscopy were used to study the formation of surface impurities on varying nickel content NMC materials in the presence of CO_2 and water. The composition of surface impurities formed in ambient air exposure was found to be dependent on water concentration and the percentage of different crystal planes. These results indicating variable surface reactivity point to a strategy to mitigate battery performance degradation through precise control of the dominant surfaces in NMC materials.



ADVANCED MATERIALS AND MANUFACTURING FOR EXTREME ENVIRONMENTS

INL recognizes the need for new materials that can perform in extreme environments, such as radiation, high temperature, corrosive, high strain-rate deformation, or explosive environments. Advanced manufacturing techniques are needed to develop many of these materials, but these techniques have not yet been applied broadly to materials for new nuclear fuels for advanced reactor designs, lightweight materials for extreme environments, or advanced survivability or protective materials. In addition, the capabilities to strategically design materials for extreme environments do not currently exist. This tremendous fundamental science challenge with real-world impact is an excellent opportunity for cross-initiative coordination leveraging the multi-program capabilities at INL. By leveraging existing expertise in materials research and development for extreme environments coupled with innovation in advanced manufacturing, INL will initiate a paradigm shift to digital design and manufacturing, away from the traditional "design-build-test" approach. Advanced manufacturing allows simultaneous fabrication and process monitoring and control and it is ideally suited to support INL mission area needs for the development of new nuclear fuels for advanced reactor fuel designs, lightweight materials for extreme environments, and advanced survivability/protective materials.



Advanced Manufacturing for Novel Nuclear Fuel and Structural Components

TOTAL APPROVED AMOUNT:
$1,336,150 over 3 years

PROJECT NUMBER:
18A12-105

PRINCIPAL INVESTIGATOR:
Nathan Jerred

CO-INVESTIGATORS:
Cheng Sun, INL
Daniel Schwen, INL
Michael McMurtrey, INL
Randall Scott, INL

By advancing the fundamental understanding of laser additive manufacturing, this research enables novel advanced reactor designs and fuel systems via tailored material performance and fabrication processes.

This study set out to characterize the micromechanical properties and phenomenological behavior of advanced fabricated structural steels. In particular, the effects of the laser additive manufacturing process on the microstructure, residual stress and tensile properties, and the changes induced from varying thermal annealing treatments was evaluated. It was observed that the 'as-deposited' 316L specimens exhibited a higher yield strength and ultimate tensile strength when compared to specimens that underwent different annealing processes. An in-depth analysis of the deformation behavior and potential irradiation tolerance of additive manufactured (AM) 316L specimens, having undergone proton irradiation, was also performed. It was found that the formation of microstructure features unique to AM fabricated 316L specimens may serve as defect sinks and provide a level of radiation tolerance in comparison to wrought 316L specimens. Further, proton-irradiated AM-316L specimens and their performance in light water reactor prototypic conditions were evaluated and compared to wrought-based materials through irradiation-assisted stress corrosion cracking (IASCC) studies.

It was observed that wrought alloys were more susceptible to cracking under IASCC conditions compared to AM fabricated specimens. This effect was attributed to the hypothesis that fabrication-induced pores, acting as defect sinks, may provide a level of irradiation tolerance. The fuel of interest, a depleted uranium-zirconium alloy (dU-10%Zr) by weight percent (wt.%), was evaluated for inclusion into the additive manufacturing processes. This included using powder atomization to produce adequate feedstock materials as well as investigating different AM methodologies and geometries to facilitate the co-manufacturing of fuel and cladding materials. Different approaches were investigated to generate a fuel structure with a fabricated internal porosity. To test and evaluate the effectiveness of these methodologies, 316L stainless steel was used as a surrogate for dU-10Zr (wt.%). It was found that varying AM build parameters could lead to internal void structures that may be larger than necessary. Further, a laser modulation technique to create micro-pores in build layers exhibited promise in generating an intentional porous structure, but continued development in this area is needed.

TALENT PIPELINE:

- Ching-Heng Shiau, student at Texas A&M University
- Fei Teng, student at Oregon State University
- Panayotis Manganaris, student at University of Cincinnati
- Rigel Hanbury, student at University of Michigan
- Sudipta Biswas, INL postdoc

PUBLICATIONS:

Shiau, C.-H., M. McMurtrey, R. O'Brien, N. Jerred, R. Scott, J. Lu, X. Zhang, Y. Wang, L. Shao, and C. Sun, "Deformation behavior and irradiation tolerance of additively manufactured 316L stainless steel," *Materials and Design*, 204 (2021) 109644.

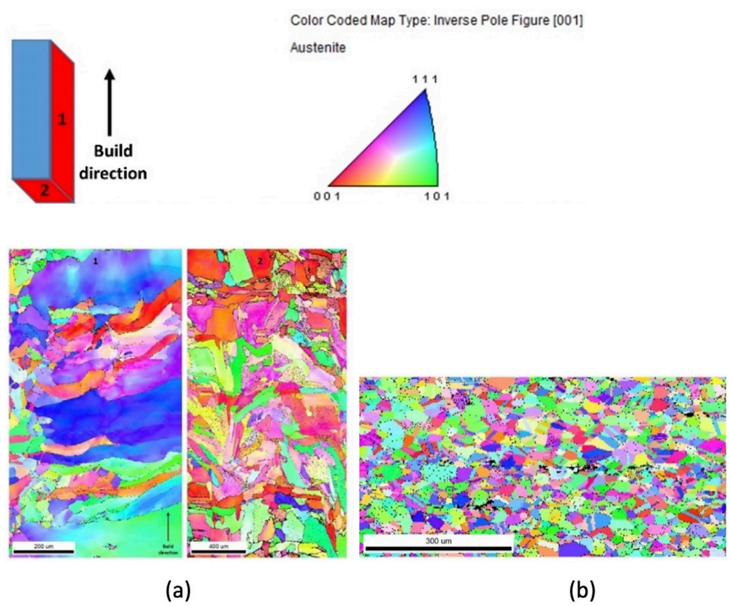
Sun, C., Y. Wang, M. McMurtrey, N. Jerred, F. Liou, and J. Li, "Additive manufacturing for energy: A review," *Applied Energy* 282 (2021) 116041.

Zhang, X., M. McMurtry, L. Wang, R. O'Brien, C.-H. Shiau, Y. Wang, R. Scott, Y. Ren, and C. Sun, "Evolution of microstructure, residual stress, and tensile properties of additively manufactured stainless steel under heat treatments," *Journal of The Minerals, Metals & Materials Society* (JOM) 72 (2020) 4167.

McMurtry, M., C. Sun, R. E. Rupp, C.-H. Shiau, R. Hanbury, N. Jerred, and R. O'Brien, "Investigation of the irradiation effects in additively manufactured 316L steel resulting in decreased irradiation assisted stress corrosion cracking susceptibility," *Journal of Nuclear Materials: Special Edition on Additive Manufacturing* 545 (2021) 152739.

PRESENTATION:

McMurtrey, M., G. Was, and X. Lou, "SCC and IASCC of printed stainless steel," *The Minerals, Metals & Materials Society Exhibition 2020*, San Diego, CA, USA, Feb. 23–27, 2020.



Microstructure of AM-316L and wrought 316L detailed using electron backscatter diffraction characterization: (a) AM-316L specimen with the build direction being shown and (b) the wrought 316L specimen.

Intelligent Additive Manufacturing

TOTAL APPROVED AMOUNT:
$1,287,038 over 3 years

PROJECT NUMBER:
18P37-030

PRINCIPAL INVESTIGATOR:
Alexander Abboud

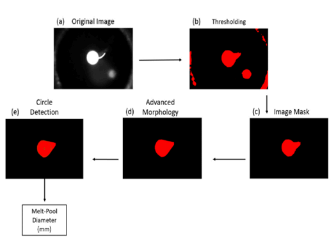
CO-INVESTIGATORS:
Michael McMurtrey, INL
Byron Pipes, Purdue University
Marco Schoen, Idaho State University
Min Long, Boise State University

Correlating additive manufacturing process parameters to the microstructure formation enables intelligent manufacturing design and reduced part variability.

This project sought to develop a better understanding of process parameters and to optimally model and control process parameters to intelligently manufacture materials and components. By creating parts with more uniform build properties, the completed research helps fill a gap to allow for additive manufacturing to become more mainstream in industries like the energy industry. Previous additive manufacturing builds contained variability in the characteristics in printed parts due to poor process control. The completed work used a combination of modeling and simulation with process monitoring and control to minimize variability in printed properties and allow for more intelligent selection of final properties for the build part. The initial material studied was stainless steel (primarily 316L), which is a well-characterized high-temperature material used in nuclear and other energy applications. The researchers developed a laser control system based on feedback from an optical camera monitoring the melt pool. Three models were developed—a computational fluid dynamics model, a phase-field model, and a FEM. These models feed information between them for accurate thermal history, microstructure development, and melt pool behavior. Significant progress was made in monitoring the effects of process parameters on final print properties, which will provide validation for the models and an increased understanding in the relation between process parameters and printed properties. The melt pool proportional–integral–derivative controller effectively maintained consistent build dimensions. An infrared camera was used to measure the temperature and should be useful in future efforts. Improving the melt pool controller with machine learning methods was also studied.

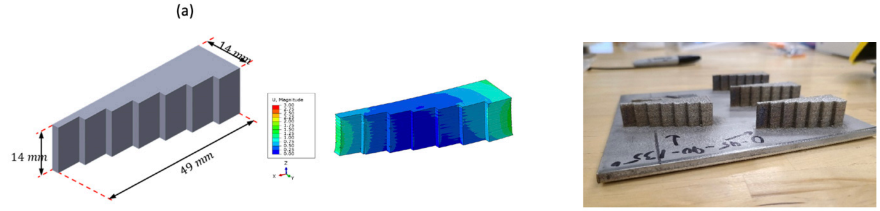
TALENT PIPELINE:

- Andy Lau, student at Boise State University
- Anthony Favaloro, postdoc at Purdue University
- Asa Monson, student at Idaho State University
- Eduardo Barocio, student at Boise State University
- Golam Jama, student at Idaho State University
- Kanan Chowdhury, student at Idaho State University



(a)

(a)



(b) (c) (d)

(a) The melt pool controller adjusts laser power feedback to maintain melt pool size, (b) and (c) multi-scale modeling and simulation using MOOSE, FEMs, and computational fluid dynamics (CFD), and (d) test parts validating models and demonstrating proof-of-concept.

Three-Dimensional Electrochemical Manufacturing and Sensing

TOTAL APPROVED AMOUNT:
$1,081,850 over 3 years

PROJECT NUMBER:
19A39-130

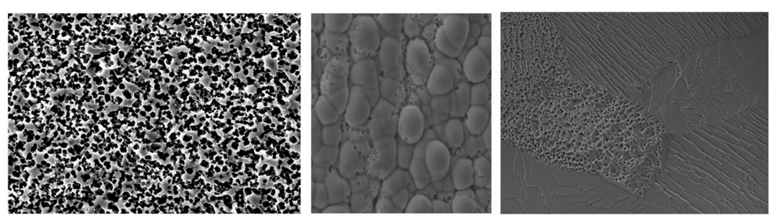
PRINCIPAL INVESTIGATOR:
Junhua Jiang

CO-INVESTIGATORS:
Congjian Wang, INL
Prabhat Tripathy, INL
Robert Mariani, INL
Haiyan Zhao, University of Idaho
Yaqiao Wu, Boise State University

COLLABORATOR:
Feng Research Labs, LLC

Novel technology enables multi-scale manufacturing of complex components and materials with low cost and low waste.

The U.S. manufacturing sector uses 25% of the nation's energy and has an annual energy bill of more than $130 billion. Advancements in manufacturing impact the energy efficiency of products used throughout the economy. Innovative manufacturing has great potential to strengthen the U.S. economy and national security, produce high income jobs, and generate technological innovation—driving long-term economic prosperity and growth. Currently, a range of advanced manufacturing technologies are under investigation for different applications. This research developed a unique three-dimensional electrochemical manufacturing and sensing (3DEMS) technology for primary advanced materials manufacturing application. It enables the fabrication of components with complicated shapes and varying dimensions, as well as controls of their chemical form, composition, and microstructures at different scales (from atomic to macro). This technology manufactures under relatively milder conditions compared to traditional technologies. The 3DEMS is also a potential sensing technology for the online monitoring of manufacturing processes and remote detection of hazardous contaminants and can also be used to fabricate advanced sensors. Furthermore, the 3DEMS is potentially scalable for mass production or can be minimized for nano-to-micro/macro machining. Specific scientific and technical accomplishments from the research include: (1) integration of a full 3DEMS system; (2) manufacturing of surrogate aluminum and aluminum-zirconium alloys with even gradient microstructures through the development of novel electrochemical processes; (3) advanced manufacturing of nuclear coatings; (4) atom irradiation for accelerated evaluation of nuclear fuels and materials; (5) electrolytic metal-atoms enabled etching and nanostructures/ heterostructures; and (6) electrochemical sensing of simulated fission products.



(a) (b) (c)

Scanning electron microscope image of (a) porous aluminum on copper, (b) aluminum-zirconium on copper, and (c) etched polycrystalline palladium.

PUBLICATIONS:

Shi, M., J. Jiang, and H. Zhao, "Electrodeposition of aluminum in the 1-ethyl-3-methylimidazolium tetrachloroaluminate ionic liquid," *Electrochemica* 2 (2021) 185–196.

Jiang, J., and C. Wang, "Review—Electrolytic metal-atoms enabled manufacturing of nanostructured sensor electrodes," *Journal of the Electrochemical Society*, 167 (2020) 037521.

Jiang, J., "Copper-assisted etching of gold through electrochemical deposition and dissolution of copper in ionic liquids," *Journal of the Electrochemical Society* 166 (2019) D940–D945.

Wu, X., J. Jiang, C. Wang, J. Liu, Y. Pu, A. Ragauskas, S. Li, and B. Yang, "Lignin-derived electrochemical energy materials and systems," *Biofuels, Bioproducts & Biorefining* 14 (2020) 650–672.

Jiang, J., and C. Wang, "Room-temperature electrodeposition of aluminum coating from 1-ethyl-3-methylimidazolium tetrachloroaluminate based ionic liquid bath," *Transactions of the American Nuclear Society*, 122 (2020) 257–258.

Jiang, J., and C. Wang, "Towards atomic irradiation and examination of nuclear materials," *Transactions of the American Nuclear Society* 123 (2020) 500–502.

Jiang, J., "Fabrication of uniform nanoparticulate gold through potential-modulated electrochemical deposition and dissolution of silver in ionic liquids," *Journal of the Electrochemical Society* 166 (2019) E521–E525.

TALENT PIPELINE:

- Meng Shi, student at University of Idaho

INTELLECTUAL PROPERTY:

Jiang, J., and R. Mariani, "3D electrodeposition systems and methods of manufacturing using such systems," U.S. Patent Application, PCT/US 19/65,395 (2019).

Jiang, J., and R. Mariani, "3D electrochemical manufacturing and sensing system and related methods," U.S. Patent Application, PCT/US 19/65,399 (2019).

Jiang, J., H. Zhao, and S. Meng, "Methods and systems for electrochemical deposition of metal from ionic liquids including imidazolium tetrahalo-metallates," U.S. Patent Application, PCT/US 17/303,945 (2021).

Jiang, J., "Electrochemically modulated molten salt reactor," U.S. Patent 11,069,449 B2 (July 20, 2021).

Jiang, J., "Flow through liquid metal cooled molten salt reactors," U.S. Patent Application, PCT/US 17/392,599 (2021).

AWARD:

DOE Energy I-Corps certificate under the financial support of DOE Energy I-Corps program ($75000), based on one innovation obtained through this project.

PRESENTATIONS:

Jiang, J., "Advanced electrochemical manufacturing of mixed dimensional heterostructures," Control #330283, *2020 Virtual Materials Research Society Spring Meeting*, Nov. 27–Dec. 4, 2020.

Jiang, J., and C. Wang, "Electrolytic metal atoms enabled manufacturing of nanoporous structures," (invited talk), Abstract #L07-2800, *237th Electrochemical Society Meeting with International Meeting on Chemical Sensors (IMCS) 2020*, Montreal, Quebec, Canada, May 10–14, 2020.

Jiang, J., and C. Wang, "Electrochemical detection using nanoporous structures," #IMCS 04-2224, *237th Electrochemical Society Meeting with International Meeting on Chemical Sensors (IMCS) 2020*, Montreal, Quebec, Canada, May 10–14, 2020.

Jiang, J., and C. Wang, "Room-temperature electrodeposition of aluminum coating from 1-ethyl-3-methylimidazolium tetrachloroaluminate based ionic liquid bath," #32285, *2020 American Nuclear Society Annual Meeting*, Virtual, Jun. 8–11, 2020.

Jiang, J., and R. Mariani, "Microscale surface nanostructuration and catalysis of metallic electrodes through electrochemical deposition and dissolution in ionic liquids," *Abstract #M03-2104, 235th Electrochemical Society Meeting*, Dallas, TX, USA, May 26–31, 2019.

Jiang, J., and M. Kerr, "Foreign metal-ion tuned electrochemical processes in ionic liquids for metal surface treatment," *236th Electrochemical Society Meeting*, Atlanta, GA, USA, Oct. 13–17, 2019.

Meng, S., H. Zhao, and J. Jiang, "Stability investigations of [EMIm] AlCl4 ionic liquid," Abstract #125586, *236th Electrochemical Society Meeting*, Atlanta, GA, USA, Oct. 13–17, 2019.

Jiang, J., Y. Wang, J. Yu, and G. Cao, "Comparative corrosion studies of structural alloys in molten fluoride and chloride salts," *Corrosion 2021*, Salt Lake City, UT, USA, Apr. 18–22, 2021.

Jiang, J., and C. Wang, "Temperature-dependent structural transformations of electrodeposited Al from acidic imidazolium-based tetrachloroaluminate ionic liquid," *PRiME 2020*, Honolulu, HI, USA, Oct. 4–9, 2020.

Jiang, J., J. Stempien, and Y. Wu, "Comparative oxidation studies of glassy carbon and nuclear graphite in dry air," *2020 American Nuclear Society Winter Meeting & Expo*, Chicago, IL, USA, Nov. 15–19, 2020.

Jiang, J., and C. Wang, "Towards atom irradiation and examination of nuclear materials," *2020 American Nuclear Society Winter Meeting & Expo*, Chicago, IL, USA, Nov. 15–19, 2020.

Design and Evaluation of a Nuclear Pumped Laser Detector for Reactor Power Indication

TOTAL APPROVED AMOUNT:
$776,129 over 3 years

PROJECT NUMBER:
19A39-146

PRINCIPAL INVESTIGATOR:
Andy Beasley

CO-INVESTIGATORS:
Adrian Wagner, INL
Scott Watson, INL
Mark Prelas, University of Missouri

COLLABORATOR:
Purdue University

New crystal development process provides pathway to produce laser crystals faster and cheaper than previously possible.

Measuring reactor and experiment power over time in test reactors, especially transient reactors, is an existing challenge. The emergence of autonomous small or micro-reactor systems will require accurate active power measurement that may be distributed at several axial and radial locations across the core. Flux and fission wires can be used to measure total fluence in the specimen in-reactor experiments but cannot provide power history or online data to intelligent controls or autonomous applications. Ion and fission chambers are typically too large to be used within small systems and cannot operate accurately at the power range seen during transient operations. A new instrument is needed to measure reactor and specimen power.

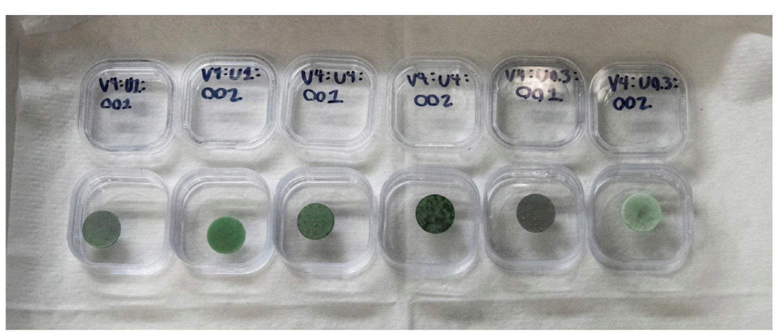
Flux in an operating nuclear reactor can be used to pump yttrium aluminum garnet (YAG) lasers. The power output from the laser is proportional to reactor power with a very fast response time. Doping the YAG crystal with uranium or plutonium allows thermal or fast neutron flux to pump the laser. By measuring the difference between undoped and doped laser output, reactor power and thermal/fast neutron flux can be determined over the range of reactor power of interest to researchers.

This research resulted in new methodologies to develop polycrystalline undoped and uranium doped YAG crystals. A new additive manufacturing process was developed to produce zirconium dioxide doped with 10% uranium dioxide, which was necessary as part of the YAG crystal manufacturing process. The YAG crystals were sized to give proper output over the expected flux for steady-state and transient reactor operations in the Neutron Radiography and Transient Reactor Test reactors, but larger crystals are achievable. This new process can produce laser crystals much faster and cheaper than the typical single crystal growing method.

TALENT PIPELINE:

- Brian Bettes, student at Purdue University
- Patrick Moo, student at University of Florida

INTELLECTUAL PROPERTY:
O'Brien, R., and A. Beasley, "Solid state nuclear pumped lasing sensors for in-pile reactor power and flux measurement, direct energy conversion, and related methods," U.S. Patent Application No. WO 2021/141882 A1 (July 15, 2021).



Polycrystalline uranium YAG samples.

Coupling of Spark Plasma Sintering with Advanced Modeling to Enable Process Scale-up

TOTAL APPROVED AMOUNT:
$1,250,000 over 3 years

PROJECT NUMBER:
19P45-031

PRINCIPAL INVESTIGATOR:
Stephanie Pitts

CO-INVESTIGATORS:
Al Casagranda, INL
Casey Icenhour, INL
Dennis Tucker, INL
Jorgen Rufner, INL
Larry Aagesen, INL
Troy Holland, INL
R. Edwin García, Purdue University
Lucas Robinson, Purdue University

Developing process-informed design through multiphysics, multi-scale modeling and simulation enables researchers to determine process-structure-property-performance correlations.

The goal of this research project was to develop a high-fidelity, multi-scale, multiphysics modeling and simulation code application for spark plasma sintering (SPS), which is valued for using less energy compared to conventional sintering, fabricating near net shape parts, and producing parts with nano-sized microstructures. Accurate modeling and simulation tools specific to SPS are needed to predict the influence of the coupled electrical, thermal, and mechanical physical processes used to create parts from metallic and ceramic micron and nano-sized powders. Voltage, temperature, and pressure are transmitted to the powder through steel rams and graphite tooling, causing the powder to undergo rapid evolution and densification. In this project, the full manufacturing apparatus was also modeled to investigate the role of the rams and tooling on the powder microstructure evolution. These two different length scales separate to an engineering-scale simulation that encompasses the full geometry of the apparatus and a set of microstructure evolution models of the sintering powder. The key outcomes of this project include two significant phase-field model developments, enhanced electro-thermal contact models, an initial multi-scale electro-thermal coupled model prototype, and the collection of a suite of experimental data to inform model development. These multiphysics simulation advances and code developments advance the ability to simulate and study other advanced manufacturing techniques, such as including laser welding and additive manufacturing applications.

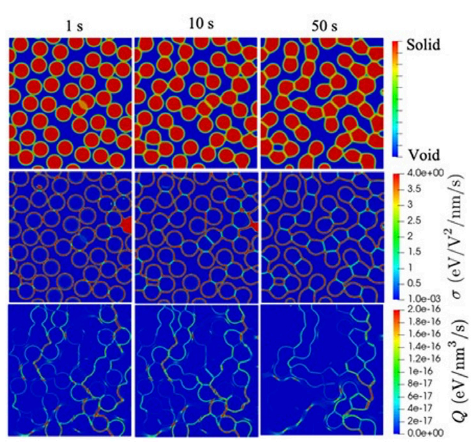
TALENT PIPELINE:

- Casey Icenhour, student at North Carolina State University
- Lucas Robinson, student at Purdue University
- Spencer Doran, student degree at Oregon State University

INTELLECTUAL PROPERTY:

Contributions to MOOSE Electromagnetic Module for enhancement of interface models and multiphysics verification and validation available on https://github.com.

Contributions to the MOOSE Application Library for Advanced Manufacturing Utilities (MALAMUTE) modeling and simulation tool available on https://hpcgitlab.hpc.inl.gov.



Microstructural evolution in a compact of yttrium oxide particles simulated using electro-thermal phase-field model. Top row: microstructure, with solid in red, void in blue, and interfaces between particles in yellow. Middle row: local electrical conductivity showing highest conductivity at the interfaces. Bottom row: heat generated during the evolution showing highest temperatures along connected interfaces.

Transient Spectrokinetic Reactor to Accelerate Development of Advanced Manufacturing Materials

TOTAL APPROVED AMOUNT:
$167,000 over 2 years

PROJECT NUMBER:
20A1047-015

PRINCIPAL INVESTIGATOR:
Rebecca Fushimi

CO-INVESTIGATORS:
James Pittman, INL
Yixiao Wang, INL
Zachary Thompson, INL

COLLABORATORS:
Idaho State University
Mithra Technologies Inc.

A novel spectrokinetic reactor was developed that couples spectral specificity with microkinetic detail to provide researchers with new methods to investigate fundamental questions of complex materials.

Manufacturing essential chemicals—ammonia, ethylene, methanol, etc.—can be more energy efficient with advanced materials such as catalysts, electrocatalysts, sorbents, and oxidation-reduction agents. These manufacturing processes take place in extreme environments and small changes in conditions can create a large impact on performance. The kinetic performance characteristics (activity and selectivity) of these materials are determined by the composition and structure of a metal/metal oxide surface, a fundamental relationship that is not well understood. Thus, operando experiments where structure is determined in an operating environment are essential. However, state-of-the-art operando devices only offer coarse kinetic information and low time-resolution on the order of seconds.

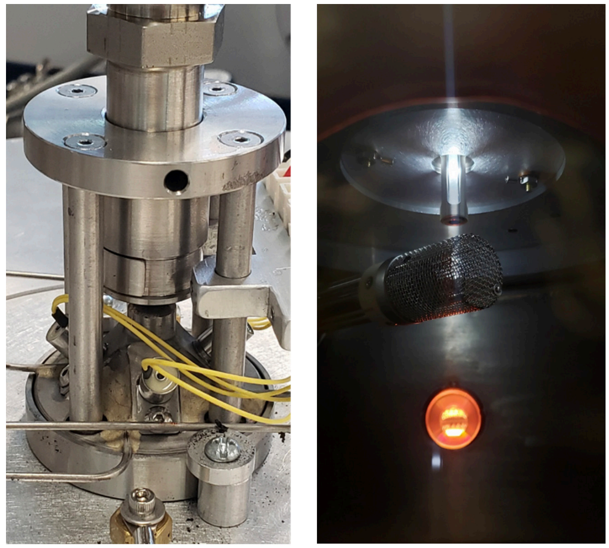
This research produced a novel spectrokinetic reactor for proof-of-concept demonstration of transient structural and kinetic characterization of materials. This work was built on the hypothesis that a diffusion reactor, similar to the TAP reactor, will provide detailed transient reaction data with millisecond time resolution while enabling resolution of the surface structural features and adsorbed molecules through Raman vibrational spectroscopy—information that is currently not accessible by any one research tool. This uniquely coupled experimental data supports advanced modeling and simulation of materials in the chemical manufacturing process as well as drive predictive tools to accelerate the design and development of materials with advanced performance properties that can reduce the intensity of the highest energy consuming chemicals.

TALENT PIPELINE:

- Bailey Vahsholtz, student at Idaho State University
- Kali Castle, student at Idaho State University
- Asa Monson, student at Idaho State University

AWARD:

Follow-on funding based on this research: Advanced Manufacturing Office (DE-FOA-0002252): Catalyst Evaluation for Deactivation and Remediation: Development of Robust Materials and Resilient Processes via Transient Measurement and Data-driven Multiscale Models ($5 M)



(a) (b)

A novel spectrokinetic reactor was developed for the study of materials under operando reaction conditions. This device provides detailed transient reaction data on the millisecond time scale in combination with changes in surface structural features: (a) gas manifold with high-speed pulse valves and (b) spectrokinetic reactor and mass spectrometer configuration in ultra-high vacuum chamber.

Probing in situ Multi-Property Evolution during Material Processing using Laser Photoacoustics

TOTAL APPROVED AMOUNT:
$215,000 over 2 years

PROJECT NUMBER:
20P1048-001

PRINCIPAL INVESTIGATOR:
Cody Dennett

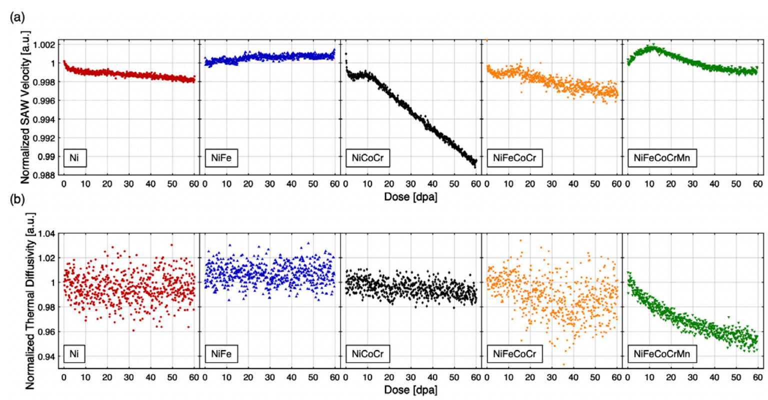
CO-INVESTIGATOR:
David Hurley, INL

COLLABORATORS:
Massachusetts Institute of Technology
University of Nebraska Lincoln
Sandia National Laboratory
University of Oxford, United Kingdom
Oak Ridge National Laboratory
University of California, Berkeley
University of California, Irvine

A new transient grating spectroscopy capability advances development of next-generation metallic material systems.

Understanding the reliability, degradation, and evolution of materials placed in combined extremes of temperature, stress, radiation, corrosives, and fatigue is fundamental to safe and secure operation of complex energy systems such as nuclear reactors. The microstructural stability of as-installed components determines whether the strength, thermal transport, and resistivity are retained throughout that component's operational lifetime. Innovation in the design and fabrication of materials used in these extreme environments is reliant on the tools used to characterize their structure and properties. This project used laser photoacoustics to observe degradation and evolution phenomena in a variety of advanced metals and alloys exposed to out-of-equilibrium conditions. These high-fidelity data will be used to optimize material compositions and fabrication pathways. The goal of developing these emerging state-of-the-art tools is to increase the utility and information density returned in single measurements.

Resulting from this research, a new experimental capability was established at INL in TGS, a laser-based method used to non-destructively measure thermal and elastic properties of materials. Using this capability, a variety of material systems in static conditions were studied, such as nickel-based concentrated solid solution alloys, nanocrystalline copper alloys, and the heat affected zones of commercial stainless steels. This new experiment has also been used for a series of in situ ion irradiation experiments carried out at the Center for Integrated Nanotechnologies using the same apparatus coupled to an ion accelerator.



Elasticity (via surface acoustic wave velocity) and thermal diffusivity of nickel-based solid solution alloys were monitored with TGS while being irradiated with nickel ions at high temperatures (left to right: nickel, nickel iron, nickel cobalt chromium, nickel iron cobalt chromium, nickel iron cobalt chromium manganese): (a) normalized change in local elasticity, which can indicate the initiation of void swelling based on the total defect population and (b) normalized change in thermal diffusivity, which can indicate that alloys with larger phonon contributions to thermal transport (for example NiFeCoCrMn) are more sensitive to radiation-induced defects than those whose primary heat carriers are electrons.

TALENT PIPELINE:

- Cody Dennett, INL
  Russell L. Heath Distinguished Postdoc

PUBLICATIONS:

Dennett, C. A., R. C. Choens, C. A. Taylor, N. M. Heckman, M. D. Ingraham, D. Robinson, B. L. Boyce, M. P. Short, and K. Hattar, "Listening to radiation damage in situ: passive and active acoustic techniques," *JOM* 72 (2020) 197–209.

Reza, A., C. A. Dennett, M. P. Short, Y. Zayachuk, J. Waite, C. Magazzeni, S. Hill, and F. Hofmann, "Non-contact, non-destructive mapping of thermal diffusivity and acoustic wave speed using transient grating spectroscopy," *Review of Scientific Instruments* 91 (2020) 054902.

Dennett, C. A., B. R. Dacus, C. M. Barr, T. Clark, H. Bei, Y. Zhang, M. P. Short, and K. Hattar, "The dynamic evolution of swelling in nickel concentrated solid solution alloys through in situ property monitoring," *Applied Materials Today* 25 (2021) 101187.

Lang, E., C. A. Dennett, N. Madden, and K. Hattar, "The in situ ion irradiation toolbox: time-resolved structure and property dynamics," *JOM* 74 (2021) 126–142.

PRESENTATIONS:

Dennett, C. A., "In situ thermoelastic property evolution of Ni-based concentrated solid solution alloys under extremes," *Annual Meeting of the Minerals, Metals, and Materials Society 2021*, Virtual, Mar. 15–18, 2021).

Dennett, C. A., "In situ observation thermoelastic material property evolution in Ni-based concentrated solid solution alloys under extreme conditions," *Materials Research Society Fall Conferenc*e, Virtual, Nov. 27–Dec. 4, 2020.

Dennett, C. A., "In situ observation of short- and long-timescale material property evolution under extreme conditions," *Materials Science and Technology Conference 2020*, Virtual, Nov. 2–6, 2020.

Dennett, C. A., "Non-destructive multi-property determination under extreme conditions with transient grating spectroscopy," *Annual Meeting of the Minerals, Metals, and Materials Society 2020*, San Diego, CA, USA, Feb. 23–27, 2020.

Ultrasonic Fatigue Lifetime Measurements: A First Step to Developing a Mechanistic Understanding

TOTAL APPROVED AMOUNT:
$125,000 over 2 years

PROJECT NUMBER:
20A1052-030

PRINCIPAL INVESTIGATOR:
Zilong Hua

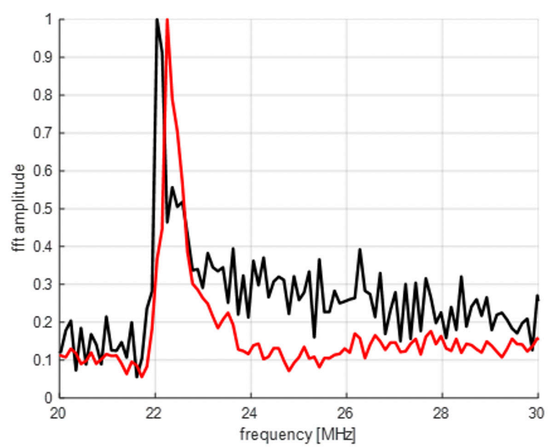
CO-INVESTIGATORS:
Cody Dennett, INL
David Hurley, INL
Michael McMurtrey, INL
Ryann Rupp, INL

COLLABORATOR:
Le Mans Université, France

A new fatigue laser ultrasonic measurement system has the potential to become a standard tool to characterize fatigue in metals, including in situ applications with complex and extreme processing conditions.

Fatigue is the initiation and growth of microscale cracks in a material related to cyclic loading. Fatigue failure always occurs in a sudden manner. Although it is a primary failure mechanism of structure materials, the understanding of fatigue is limited. Due to fatigue's stochastic nature, most fatigue models are empirically concluded from many experiments and specimens. It is extremely difficult to predict the fatigue lifetime of a single component, and preventing fatigue relies on costly safety margins.

This research explored the possibility of using a laser ultrasonic-based tool to predict fatigue life of individual components and study fatigue mechanisms. Ultrasonic waves are sensitive to microscale crack generation and growth during the fatigue process. When the ultrasonic waves encounter microcracks, wave velocity, amplitude, and frequency change correspondingly. A system capable of performing cyclic fatigue test and laser ultrasonic measurements on solid centimeter-size specimens was built, and a zero-group-velocity mode Lamb wave was investigated to study fatigue. The zero-group-velocity plate wave has a high signal to noise ratio and is insensitive to the boundary conditions. Experiments using aluminum foil revealed a relationship between the zero-group-velocity wave propagation and the microstructure change, such as microstructure defect density increase and new defects type generation.



Frequency spectrum showng zero-group-velocity plate wave changes before the test (black line) and before the final fracture (red line). Note the frequency peak shift, peak width change, and the appearance of multiple peaks near the main one.

Mitigating Irradiation-assisted Stress Corrosion Cracking by Rapid Alloy Design

TOTAL APPROVED AMOUNT:
$1,713,456 over 3 years

PROJECT NUMBER:
19A39-071

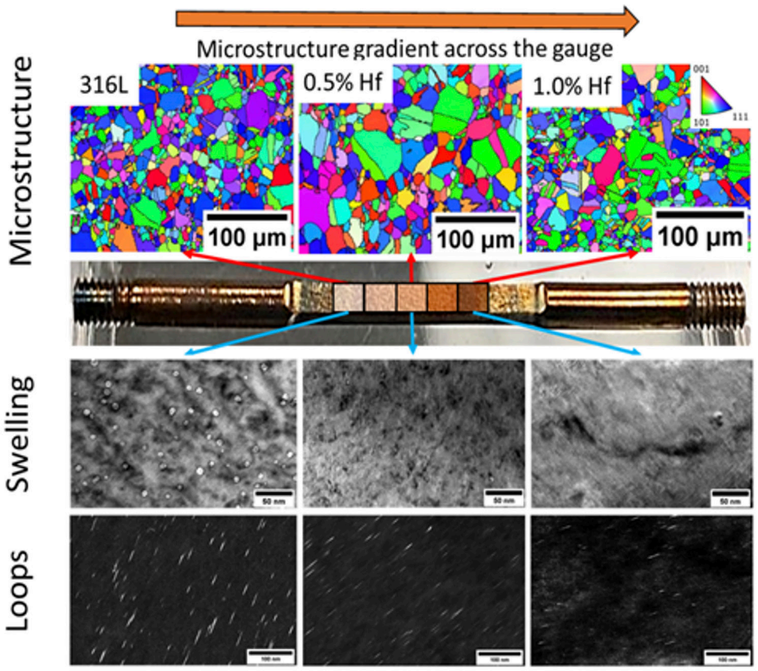
PRINCIPAL INVESTIGATOR:
Daniel Schwen

CO-INVESTIGATORS:
Lingfeng He, INL
Xiang Liu, INL
Lin Shao, Texas A&M University
Miao Song, University of Michigan
Xiaoyuan Lou, Auburn University
Yongfeng Zhang, University of Wisconsin
Zhijie Jiao, University of Michigan

COLLABORATOR:
Andresen Consulting

A rapid alloy design framework has broad applications to accelerate the design of irradiation resistant materials.

This research demonstrated a rapid alloy design framework for developing reactor structural materials. It directly addresses several challenges that lead to long and expensive development cycles of nuclear materials, including complex design space, slow manufacturing of nuclear grade samples, and expensive and time-consuming in-pile tests. A knowledge-based optimization approach was utilized in place of empirical, test-based approaches. By coupling Integrated Computational Materials Engineering, rapid prototyping and fabrication, and out-of-pile testing, material science principles can be established for materials behaviors in extreme conditions. These principles were used to guide materials optimization for improved performance in current reactors and for applications in new operation conditions and new types of reactors. The approach is demonstrated by optimization of stainless steels for improved resistance to IASCC, a critical issue concerning reactor safety. The research demonstrated the feasibility of using Integrated Computational Materials Engineering for recommendations of optimization and using advanced manufacturing for rapid prototyping and fabrication. Candidates for stainless steel additives were down-selected and gradient samples with varying additive concentrations were printed. A post-processing treatment was developed to ensure the printed material microstructure is similar to wrought material. Out-of-pile irradiations up to 50 displacements per atom were performed on the novel alloys. Characterization confirmed the trends in reducing vacancy mobility predicted by modeling and simulation.



Microstructure, radiation damage, and IASCC assessment from the compositionally graded 316L stainless steel tensile bar with selected transition metal elements, including yttrium, hafnium, zirconium, tantalum, and titanium that were fabricated by laser direct energy deposition additive manufacturing.

TALENT PIPELINE:

- Aashique Rezwan, postdoc at University of Wisconsin
- Anton Schneider, postdoc at University of Wisconsin
- Anus Manzoor, student at University of Wyoming
- Houshang Yin, student at Auburn University
- Jingfan Yang, student at Auburn University
- Laura Hawkins, student at Texas A&M University
- Sourabh Kadambi, INL postdoc

PUBLICATIONS:

Zhang, Y., A. Manzoor, C. Jiang, D. Aidhy, and D. Schwen, "A statistical approach for atomistic calculations of vacancy formation energy and chemical potentials in concentrated solid-solution alloys," *Computational Materials Science* 190 (2021) 110308.

Morgan, D., and Y. Zhang, "Comment on 'Thermal vacancies in random alloys in the single-site mean-field approximation,'" *Physical Review B* 101(13) (2020) 136101.

Manzoor, A., Y. Zhang, and D. Aidhy, "Factors affecting vacancy formation energy in Fe70Ni10Cr20 random concentrated alloy," *Computational Materials Science* 198 (2021) 110669.

Yang, J., M. Song, L. R. Hawkins, X. Liu, L. He, and X. Lou, "Effects of heat treatment on corrosion fatigue and stress corrosion crack growth of additive-manufactured Alloy 800H in high-temperature water," *Corrosion Scienc*e 191 (2021) 109739.

Yin, H., M. Song, P. Deng, L. Li, B. C. Prorok, and X. Lou, "Thermal stability and microstructural evolution of additively manufactured 316L stainless steel by laser powder bed fusion at 500-800 °C," *Additive Manufacturing* 41 (2021) 101981.

He, L., L. Hawkins, J. Yang, X. Liu, M. Song, X. Lou, Y. Zhang, L. Shao, and D. Schwen, "Advanced characterization of additively manufactured 316L stainless steel for nuclear applications," *Microscopy and Microanalysis* 27 (S1) (2021) 2160–2161.

PRESENTATIONS:

Zhang, Y., A. Manzoor, D. Aidhy, M. Song, X. Lou, and L. He, "The effect of minor additives on radiation induced segregation in austenitic steel alloys," *The Minerals, Metals & Materials Society 2020 Annual Meeting & Exhibition*, San Diego, CA, USA, Feb. 23–27, 2020.

Zhang, Y., M. Song, X. Liu, L. He, D. Schwen, and X. Lou, "Improving irradiation resistance of alloys by controlling defect diffusion: A modeling perspective," *The Minerals, Metals & Materials Society 2021 Annual Meeting & Exhibition*, Virtual, Mar. 15–18, 2021.

Zhang, Y., A. Manzoor, C. Jiang, and D. Aidhy, "A Statistical Approach for Atomistic Calculations of Vacancy Formation Energy and Chemical Potentials in Concentrated Solid solution Alloys," *Materials Science & Technology Technical Meeting and Exhibition 2021*, Virtual, Oct. 17–21, 2021.

He, L., L. Hawkins, J. Yang, X. Liu, M. Song, X. Lou, Y. Zhang, L. Shao, and D. Schwen, "Advanced characterization of additively manufactured 316L stainless steel for nuclear applications," *Microscopy and Microanalysis* 27(S1) (2021) 2160–2161.

Yang, J., "Compositionally graded specimen as a high-throughput alloy screening method for evaluating irradiation-assisted stress corrosion cracking," *2021 Annual Meeting of International Cooperative Group on Environmentally-Assisted Cracking, Kingston*, Ontario, Canada, May 16–20, 2021.

Lou, X., J. Yang, X. Liu, M. Song, and L. He, "Sensitization/desensitization and carbide evolution of alloy 800H made by laser powder bed fusion," *NACE Corrosion 2021 Conference and Expo, Virtual*, Apr. 19–30, 2021.

Yang, J., X. Liu, M. Song, L. He, and X. Lou, "Sensitization and stress corrosion cracking of alloy 800H by laser powder bed fusion," *The Minerals, Metals & Materials Society 2021 Annual Meeting & Exhibition*, Virtual, Mar. 15–18, 2021.

Song, M., J. Yang, X. Liu, X. Lou, Y. Zhang, L. He, and D. Schwen, "A superb void swelling resistant type 316L stainless steel developed by additive manufacturing enabled high throughput microalloying," *The Minerals, Metals & Materials Society 2021 Annual Meeting & Exhibition*, Virtual, Mar. 15–18, 2021.

Liu, X., J. Yang, M. Song, X. Lou, Y. Zhang, L. He, and D. Schwen, "Proton irradiation induced microstructural evolution in compositionally graded type 316L stainless steel," *The Minerals, Metals & Materials Society 2021 Annual Meeting & Exhibition*, Virtual, Mar. 15–18, 2021.

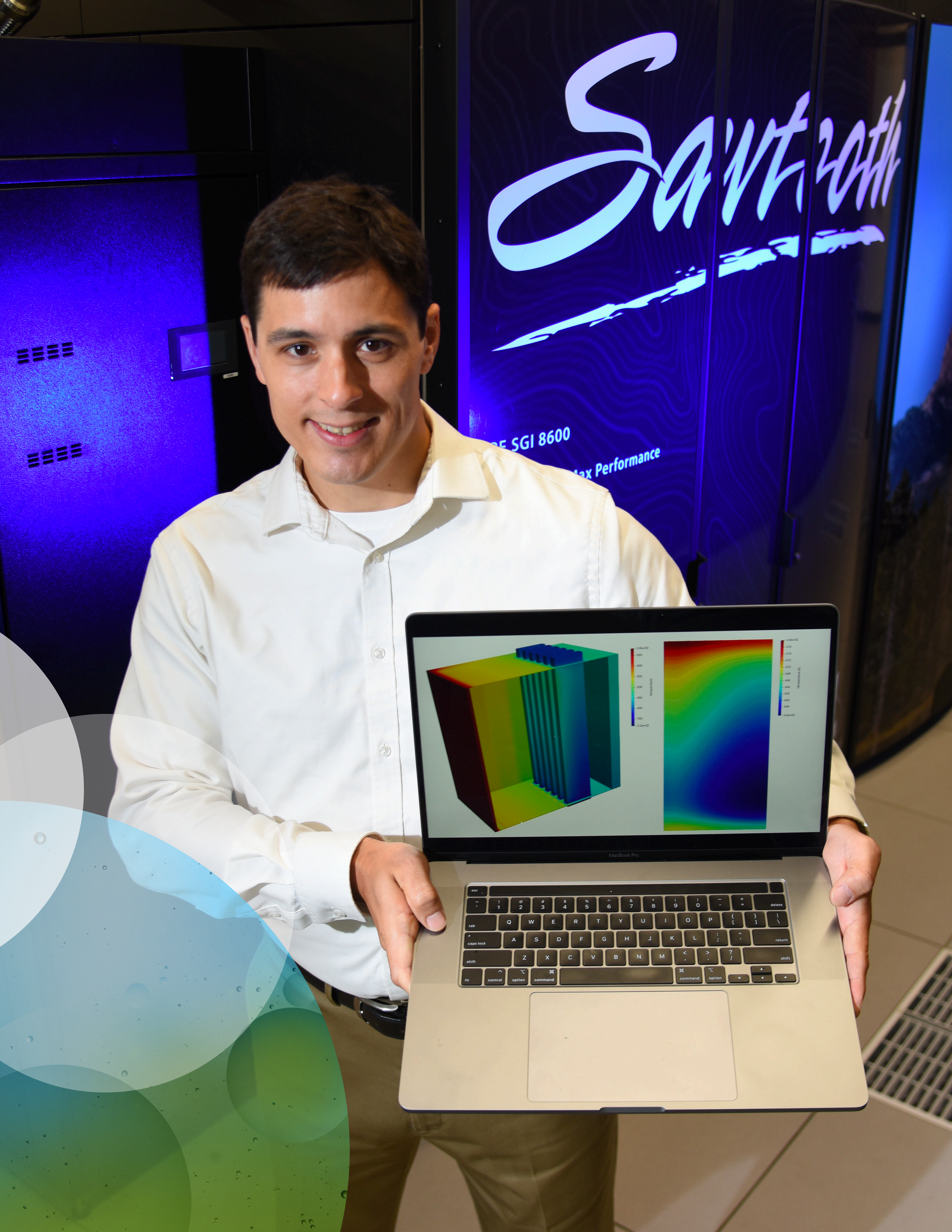
Lou, X., J. Yang, X. Liu, M. Song, L. He, Y. Zhang, and D. Schwen, "Compositionally graded bulk specimen: A high-throughput approach for nuclear alloy development and qualification," *The Minerals, Metals & Materials Society 2021 Annual Meeting & Exhibition*, Virtual, Mar. 15–18, 2021.

Yang, J., X. Liu, M. Song, L. He, Y. Zhang, and X. Lou, "Irradiation-assisted stress corrosion cracking (IASCC) of austenitic stainless steels with oversized solutes in high temperature water," *The Minerals, Metals & Materials Society 2021 Annual Meeting & Exhibition*, Virtual, Mar. 15–18, 2021.

SECURE AND RESILIENT CYBER-PHYSICAL SYSTEMS

The cyber-physical systems within the nation's critical civilian infrastructure and military systems must be secured against an increasingly complex and dynamic set of cyber threats and be resilient to a variety of hazards and environmental changes. Traditional information assurance approaches for securing computer technology are necessary, but not sufficient, to provide functional assurance within critical infrastructures, their complex supply chain of components, and their networks of integrators and operators. Cyber adversaries are increasing in size and complexity, vulnerability discovery and exploitation is being automated (often faster than mitigations can be developed), and in many cases, adversaries are using safety or security features as part of their attack. In this environment, neither security nor resiliency is achieved with "add-on" technologies or traditional perimeter defenses. Transformational approaches based on engineering principles, formal validation, and consequence management are needed to address the true risks of critical systems throughout their lifecycle.

Leveraging its core capabilities, INL drives both a culture and technology change in critical systems engineering to infuse security and resiliency throughout an infrastructure's lifecycle. This includes the science and engineering capabilities for proactive design, implementation, and operation of secure and resilient systems. Future systems and technologies must proactively eliminate weakness in designs and minimize high-consequence risks from sophisticated, persistent, and co-adaptive adversaries; and they must rapidly detect and respond to new cyberattacks, as well as physical threats and environmental changes. The nation also has a fraction of the necessary expertise to defend existing vulnerable systems—much less address the fundamental flaws in present infrastructure security and resiliency approaches. These workforce gaps, coupled with fundamental challenges and rapidly changing technology, make the spectrum and pace of infrastructure RD&D particularly daunting and increases the importance of INL's strategic partnerships for research-enabled work force development initiatives with universities, colleges, and science, technology, engineering, and math programs.


Sawtooth
SGI 8600
Performance

Novel Methods to Produce an Argon-37 Standard

TOTAL APPROVED AMOUNT:
$700,592 over 3 years

PROJECT NUMBER:
18A12-076

PRINCIPAL INVESTIGATOR:
Edna Cárdenas

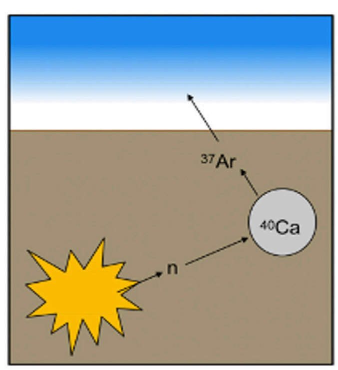
CO-INVESTIGATORS:
Ariana Foley, INL
Brian Bucher, INL
Jacob Brookhart, INL
Jessica Ward, INL
Mathew Snow, INL
Troy Robinson, INL
Jon Stoner, Idaho Accelerator Center

COLLABORATOR:
Idaho Accelerator Center

New production methods for argon-37 standards improve global capabilities for establishing evidence of unsanctioned underground nuclear detonations.

The purpose of this research was to produce a measurable quantity of radioactive argon-37 (^{37}Ar) through photonuclear production methods. The detection of ^{37}Ar above background levels uniquely and clearly indicates the underground detonation of a nuclear device. Fission neutrons generated from a nuclear blast interact with calcium present in rocks and soil in the underground environment. The interaction of these fission neutrons with calcium-40 (^{40}Ca) produces ^{37}Ar, through a ^{40}Ca(neutron, alpha) ^{37}Ar reaction. The gas then seeps through the underground environment to the surface for detection by on-site inspectors. Inspections are performed to aid in the regulation of nuclear non-proliferation treaties, such as the Comprehensive Nuclear-Test-Ban treaty. ^{37}Ar's half-life of approximately 35 days means that detection is possible long after the detonation, making its identification valuable during on-site inspections.

Successful production of ^{37}Ar on a small scale would provide proof-of-principle leading to opportunities for yet-to-be realized large scale sample production. An ^{37}Ar standard would give on-site inspectors the ability to calibrate their detectors and measure detector efficiency, thus providing higher accuracy in quantifying detected ^{37}Ar. An ^{37}Ar standard would also provide authentic samples to test novel detectors and improve models predicting the diffusion of ^{37}Ar in an underground environment. Current research into the production of ^{37}Ar utilizes neutrons generated from nuclear reactors on samples containing ^{40}Ca. The research in this project studied photonuclear production of ^{37}Ar through photon bombardment on potassium-39, ^{40}Ca, and ^{38}Ar using electron linear accelerators. Linear accelerators are more readily available than nuclear reactors used for experiments and have a wide range of power output, allowing for the optimization of ^{37}Ar production and potentially providing a novel production pathway. This project advanced research in security solutions that prevent, detect, and counter nuclear and radiological threats.



Fission neutrons interacting with the calcium in rocks and soil generate ^{37}Ar at a nuclear detonation site. The ^{37}Ar seeps to the surface where it can be detected by sensors.

TALENT PIPELINE:

- Ariana Foley, student at Oregon State University
- Edna Cárdenas, INL postdoc
- Jessica Ward, student at University of Idaho

PRESENTATIONS:

Ward, J. L., M. S. Snow, and E.S. Cárdenas, "Novel methods in producing ^{37}Ar standards," *64th Radiobioassay & Radiochemical Measurements Conference (RRMC)*, Santa Fe, NM, USA, Oct. 27–Nov. 1, 2019.

Cárdenas, E. S., A. A. Foley, B. M. Bucher, J. L. Brookhart, T. A. Robinson, M. S. Snow, and J. L. Ward, "^{37}Ar standard – Improves radionuclide monitoring tools," *Presented to NA-22 leadership in Washington D.C.*, Oct. 2019.

Foley, A. A., B. M. Bucher, J. L. Brookhart, T. A. Robinson, M. S. Snow, J. L. Ward, and E. S. Cárdenas, "Novel photonuclear methods to produce an argon-37 standard", *American Nuclear Society Annual Winter Meeting 2020*, Virtual, Nov. 16–19, 2020.

Robust Insulation for Resilient Transformers against an Electromagnetic Pulse or Geomagnetic Disturbance

TOTAL APPROVED AMOUNT:
$1,412,440 over 3 years

PROJECT NUMBER:
18P37-033

PRINCIPAL INVESTIGATOR:
Bjorn Vaagensmith

CO-INVESTIGATORS:
Gorakh Pawar, INL
Jesse Reeves, INL

COLLABORATOR:
University of Utah

Improving temperature tolerance of the electrical insulation used in transformers will increase the electrical grid resilience against geomagnetic disturbances and electromagnetic pulses.

Power transformers, a vital grid component, are vulnerable to premature failure during and after geomagnetic disturbances that cause elevated temperatures and voltage excursions that compromise internal insulation. Current state-of-the-art insulations use organic polymers or micro-fibers imbedded in a temperature sensitive binding matrix for structural stability, which limits the thermal tolerance. A new very high-temperature tolerant insulation is needed. This research investigated novel insulation materials with enhanced thermal properties using engineered silica fibers fabricated via electrospinning. These unique electrospun submicron silica fibers were converted into prototype felt insulation for use in new compact transformers for the electrical grid and specialty applications and to provide greater resilience against temperature excursions and overvoltage during a geomagnetic disturbance.

Mats composed of silica fibers ranging in diameter from 0.16–4.12 µm were fabricated with thicknesses up to 200 µm. These mats were resilient to temperature stresses up to 1100°C (heated for 1 hour), electrical resistivity of 1.8×10^{14} Ω-cm, and break down voltages of 6.5 MV/m (for a dry test). Molecular dynamic simulations indicated that the molecular bonds begin to break apart at a significantly increased rate around 677–727°C. These results approached or exceeded the original goals to survive temperatures up to 450°C, electrical resistivity of 10^{15} Ω-cm and a break down voltage of 10 MV/m. Increasing the mat density increased the electrical resistivity to 2.6×10^{14} Ω-cm, and further research has the potential to achieve 10^{15} Ω-cm.

Conversely, the tensile strength of the mats was three orders of magnitude weaker (500 kPa) compared to commercial insulation products, such as Kraft paper (50 MPa). After the sol-gel cured for several months, the tensile strength increased to 1–5 MPa. Alternative fabrication methods were explored to increase the tensile strength; more research is needed to reach the goal.

Examples of silica fiber mats developed under this research.

TALENT PIPELINE:

- Ezekiel Current, student at Salt Lake Community College
- Jacob Ulrich, student at Iowa State University
- Jim Greene, student at Brigham Young University-Idaho
- Kushalta Paudel, student at Boise State University
- Rajni Chahal, student at University of Texas at Arlington
- Schuyler McFall, student at University of Utah
- Tony Tuss, student at Boise State University
- William Serrano-Garcia, student at University of South Florida

PRESENTATION:

Pawar, G., B. Vaagensmith, B. Isaac, and T. Borders, "Design and optimization of the advanced silica nanofiber insulator material properties via coarse-grain molecular dynamics simulation method," *2019 Materials Research Society Spring Meeting & Exhibit*, Phoenix, AZ, USA, Apr. 22–26, 2019.

INTELLECTUAL PROPERTY:

Reeves, J., and B. Vaagensmith, "Silica nanofiber materials, articles including such materials, and related methods," U.S. Patent Application No. 16/430,478.

Boron Suboxide and Refractory Transition Metal Boride Fabrication by Spark Plasma Sintering

TOTAL APPROVED AMOUNT:
$1,492,958 over 3 years

PROJECT NUMBER:
18A12-116

PRINCIPAL INVESTIGATOR:
Joshua Kane

CO-INVESTIGATORS:
Henry Chu, INL
Jeffrey Anderson, INL
Thomas Lillo, INL
Troy Holland, INL

COLLABORATORS:
Brookhaven National Laboratory
Carl Zeiss Microscopy
Oak Ridge National Laboratory

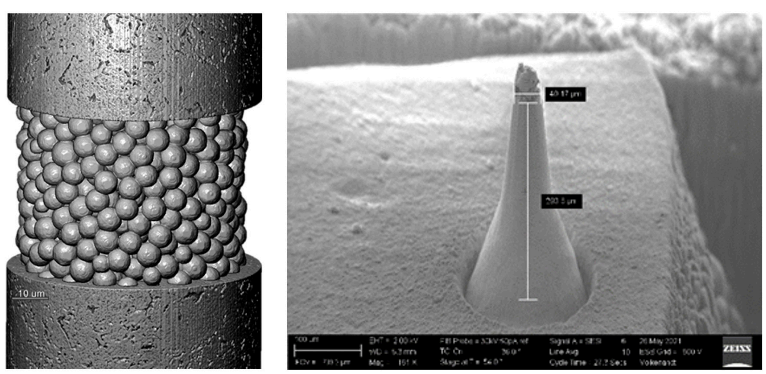
Advancing spark plasma sintering toward industrial scale by increasing understanding of the sintering kinetics and fundamental phenomena.

Boron suboxide and transition metal borides, such as tungsten tetraboride, were identified by several U.S. Department of Defense branches as potential next-generation ceramic armor materials. These materials have a range of outstanding physical and chemical properties applicable to armor, such as a high bulk and shear moduli, hardness, thermal stability, and chemical inertness all while maintaining relatively low densities. These attributes are essential to improve on existing armor systems performance and improve the overall compatibility with the users of these armor systems. The challenges with large-scale synthesis and conventional consolidation techniques have, until recently, impeded the pathway to utilization of these materials in practical armor systems.

This project focused on utilizing SPS methods to determine the sintering characteristics and optimum sintering parameters for boron suboxide and tungsten tetraboride. SPS has long shown great promise for ceramics and refractory materials traditionally considered difficult or impossible to fabricate on an industrial scale. The kinetics of the process, however, are convoluted and difficult to ascertain, making the industrial adaptation of the technique limited. This project designed and built specialized SPS systems, including a miniature SPS, to allow experiments involving advanced in situ radiography while operating the systems. These experiments enabled the first ever SPS experiments with active interrogation to deconvolute the coupled internal (e.g., electrical, mechanical, and thermal) phenomena under SPS to understand the correct sintering kinetics. The result will enable the advancement of SPS science and allow the process to be optimized for scale-up.

TALENT PIPELINE:

- Amanda Smolinski, student at Idaho State University
- Jeffrey Anderson's, student at Pennsylvania State University
- Robert Mach, student at University of Texas El Paso



(a) (b)

(a) Three-dimensional rending of SPS experiment with particles between two carbon electrodes. (b) Image of fabricated pillar.

An Innovative Secure Millimeter Wave Machine-to-Machine Communication Network for Operating Drones

TOTAL APPROVED AMOUNT:
$1,377,500 over 3 years

PROJECT NUMBER:
19A39-023

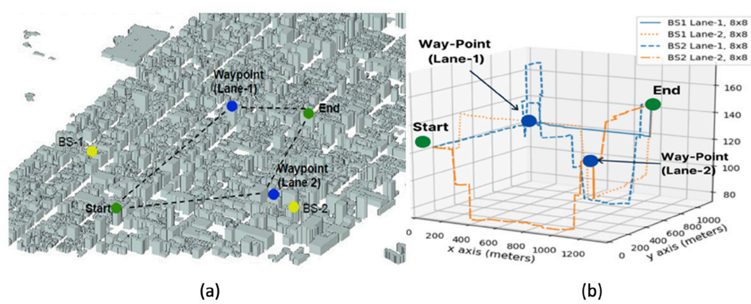
PRINCIPAL INVESTIGATOR:
Arupjyoti Bhuyan

CO-INVESTIGATORS:
Huaiyu Dai, North Carolina State University
Ismail Guvenc, North Carolina State University

This research developed a technical basis for the future research needed to mature the technology for a nationwide secure and reliable network supporting many government and public safety drones.

Researchers investigated novel approaches to improve spectral efficiency and security with 5th generation (5G) wireless networks using millimeter wave (mmWave) frequency bands to operate unmanned aerial vehicles with machine-to-machine communication. A new network approach will provide a superior alternative to existing methods with improved radio frequency coverage and resiliency against cyberattacks.

The team evaluated the security, reliability, and spectral efficiency of the proposed cellular network for unmanned aerial vehicles and drone operations under four primary experimental scenarios. The radio frequency coverage in the proposed 'drone corridor' when adding a separate set of antennas for drone coverage in the air while the conventional set of antennas continued to provide coverage on the ground was analyzed. The application of multiple-input and multiple-output access technology to increase the spectral efficiency by covering a swarm of drones with a single mmWave beam and associated radio resources was investigated. Drone groupings for non-orthogonal multiple access technology and rate-splitting multiple access technology in presence of interference were optimized. Proposed power allocation strategies for fingerprint-based authentication procedure when artificial noise is also transmitted for added security were analyzed. Pre-coders that can enhance physical layer security with channel information about the interference source were designed and evaluated. This research provided proof-of-concept to add a separate set of antennas for 5G coverage in the air for a nationwide drone corridor for 5G cellular drones. In the future, the Aerial Experimental and Research Platform for Advanced Wireless at North Carolina State University can be used to collect experimental data to further advance this research.



(a) Drone corridor in a region of East Manhattan. (b) Optimal trajectories using 8x8 arrays at the base stations BS-1 and BS-2.

PUBLICATIONS:

Maeng, S., Y. Yapici, I. Güvenç, H. Dai, and A. Bhuyan, "Precoder design for physical-layer security and authentication in massive MIMO UAV communications," *IEEE Transactions on Vehicular Technology* (2022) Ahead-of-Print. Available at: www.doi.org/10.1109/TVT.2022.3141055.

Chowdhury, M. U., İ. Güvenç, W. Saad, and A. Bhuyan, "Ensuring reliable connectivity to cellular-connected UAVs with uptilted antennas and interference coordination," *ITU Journal on Future and Evolving Technologies* (ITU J-FET) 2(2) (2021) 165–185.

Ding, H., H. Dai, I. Güvenc, and A. Bhuyan, "Outage analysis for cooperative mmWave UAV communications with beam training overhead," *IEEE Wireless Communications Letters* 10(10) (2021) 2249–2253.

Yapıcı, Y., N. Rupasinghe, İ. Güvenç, H. Dai, and A. Bhuyan, "Physical layer security for NOMA transmission in mmWave drone networks," *IEEE Transactions on Vehicular Technology* 70(4) (2021) 3568–3582.

Singh, S., H. W. Lee, T. X. Tran, M. L. Sichitiu, I. Güvenc, and A. Bhuyan, "FPV adaptation for UAV collision avoidance," *IEEE Open Journal of the Communications Society* 2 (2021) 2095–2110.

Rahmati, A., S. Hosseinalipour, Y. Yapici, X. He, I. Güvenç, H. Dai, and A. Bhuyan, "Dynamic interference management for UAV-assisted wireless networks for NOMA transmission in mmWave drone networks," *IEEE Transactions on Wireless Communications* (2021) Ahead-of-Print. Available at: www.doi.org/10.1109/TWC.2021.3114234.

Maeng, S. J., M. A. Deshmukh. I. Güvenç, H. Dai, and A. Bhuyan, "Interference analysis and mitigation for aerial IoT considering 3D antenna patters," *IEEE Transactions on Vehicular Technology* 70(1) (2021), 490–503.

Maeng, S. J., Y. Yapici, I. Güvenç, H. Dai, and A. Bhuyan, "Hybrid precoding for mmWave massive MIMO with one-bit DAC," *IEEE Communication Letters* 24(12) (2020) 2941–2945.

Rupasinghe, N., Y. Yapici, I. Güvenc, H. Dai, and A. Bhuyan, "Physical layer security for UAV communications," In: *UAV Communications for 5G and Beyond*, Wiley IEEE Press, 2021.

TALENT PIPELINE:

- Ali Rahmati, student at North Carolina State University
- Ender Ozturk, postdoc at North Carolina State University
- Harini Narasimhan, student at North Carolina State University
- S. J. Maeng, student at North Carolina State University
- Simran Singh, student at North Carolina State University
- Sri Latha Sunkara, student at North Carolina State University
- Udita Bhattacherjee, student at North Carolina State University
- Yavuz Yapici, postdoc at North Carolina State University

INTELLECTUAL PROPERTY:

Bhuyan, A., I. Güvenc, H. Dai, "An innovative secure millimeter-wave (mmWave) machine to machine (M2M) communication network for operating drones," PCT/US patent for Provisional patent 2935-P14461US (Nov 5, 2019).

PRESENTATIONS:

Du, K., O. Mujumdar, O. Ozdemir, E. Ozturk, I. Güvenc, M. L. Sichitiu, H. Dai, and A. Bhuyan, "60 GHz outdoor propagation measurements and analysis using Facebook terragraph radios," *2022 IEEE Radio & Wireless Week (RWW2022)*, Jan. 2022.

Singh, S., U. Bhattacherjee, E. Ozturk, İ. Güvenç, H. Dai, M. Sichitiu, and A, Bhuyan, "Placement of mmWave base stations for serving urban drone corridors," *2021 IEEE 93rd Vehicular Technology Conference*, Apr. 2021.

Bhuyan, A., İ. Güvenç, H. Dai, M. L. Sichitiu, S. Singh, A. Rahmati, and S.J. Maeng, "Secure 5G network for a nationwide drone corridor," *2021 IEEE Aerospace Conference*, March 2021.

Maeng, S. J., Y. Yapici, İ. Güvenç. H. Dai, and A. Bhuyan, "Power allocation for fingerprint-based phy-layer authentication with mmWave UAV networks," *IEEE International Conference on Communications* (ICC), Jun. 2021.

Rahmati, A., S. Hosseinalipour, Y. Yapici, İ. Güvenç, H. Dai, and A. Bhuyan, "Energy-efficient beamforming and power control for uplink NOMA in mmWave UAV networks," *GLOBECOM 2020 - 2020 IEEE Global Communications Conference*, Nov. 2020.

Rahmati, A., S. Hosseinalipour, I. Güvenc, H. Dai, and A. Bhuyan, "Lifetime maximization for UAV-assisted data gathering networks in the presence of jamming," *21st IEEE International Workshop on Signal Processing Advances in Wireless Communications (IEEE SPAWK)*, May 2020.

Maeng, S. J., Y. Yapıcı, I. Güvenc, H. Dai, and A. Bhuyan, "Precoder design for mmWave UAV communications with physical layer security," *21st IEEE International Workshop on Signal Processing Advances in Wireless Communications (IEEE SPAWK)*, May 2020.

Singh, S., S. L. Sunkara, I. Güvenc¸ A. Bhuyan, H. Dai, and M. L. Sichitiu, "Coverage analysis for ground and aerial users in mmWave cellular networks in urban settings," *IEEE Southeastcon*, Mar. 2020.

Singh, S., S. L. Sunkara, I. Güvenc¸ A. Bhuyan, H. Dai, and M.L. Sichitiu "Spectrum reuse among aerial and ground users in mmWave cellular networks in urban settings," *IEEE Consumer Communications & Networking Conference (CCNC)*, Jan. 2020.

Rahmati, A., Y. Yapıcı, I. Güvenc, H. Dai, and A. Bhuyan, "Interference avoidance UAV assisted networks: Joint 3D trajectory and power optimization," *IEEE Global Communication Conf. (GLOBECOM)*, Dec. 2019.

Maeng, S. J., M. A. Deshmukh, I. Güvenc, and A. Bhuyan, "Interference mitigation scheme in 3D topology IoT network with antenna radiation pattern," *IEEE Global Communication Conf. (GLOBECOM)*, Dec. 2019.

Yapıcı, Y., I. Güvenc, H. Dai, and A. Bhuyan, "Physical layer security for UAV swarm communications via protected zone," *Resilience Week*, Nov 2019.

Bhuyan, A., I. Güvenc, H. Dai, Y. Yapıcı, A. Rahmati, and S. J. Maeng, "Secure mmWave cellular network for drone communication," *IEEE Vehicular Technology Conference*, Sep. 2019.

Maeng, S. J., M. A. Deshmukh, I. Güvenc, and A. Bhuyan, "Interference mitigation scheme in 3D topology IoT network with antenna radiation pattern," *IEEE Vehicular Technology Conferenc*e, Sep. 2019.

Yapıcı, Y., S. J. Maeng, I. Güvenc, H. Dai, and A. Bhuyan, "SLNR based precoding for one-bit quantized massive MIMO in mmWave communications," *IEEE International Conference on Communications (ICC)*, May 2019.

Rahmati, A., Y. Yapıcı, N. Rupasinghe, I. Güvenc, H. Dai, and A. Bhuyan, "Energy efficiency of RSMA and NOMA in cellular-connected mmWave UAV networks," *IEEE International Conference Communications* (ICC), May 2019.

Rupasinghe, N., Y. Yapıcı, I. Güvenc, H. Dai, and A. Bhuyan, "Enhancing physical layer security for NOMA transmission in mmWave drone networks," *IEEE Asilomar Conference Signals, Systems, and Computer*, Oct. 2018.

Secure Millimeter Wave Spectrum Sharing with Autonomous Beam Scheduling

TOTAL APPROVED AMOUNT:
$890,000 over 2 years

PROJECT NUMBER:
20A44-058

PRINCIPAL INVESTIGATOR:
Arupjyoti Bhuyan

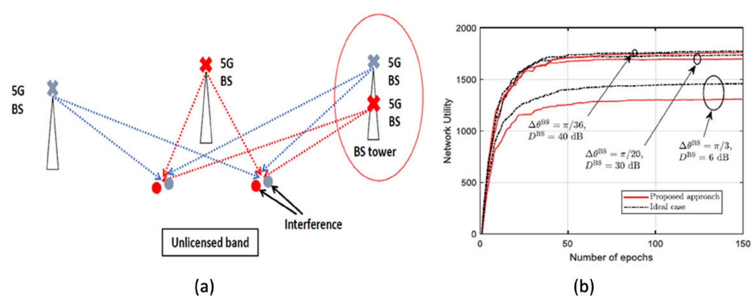
CO-INVESTIGATORS:
Jacobus Van der Merwe, University of Utah
Mingyue Ji, University of Utah
Sneha Kasera, University of Utah

This research introduced a distributed scheduler for cellular base stations that can be further matured as an innovative technology for the emerging field of 5G spectrum sharing without requiring centralized control.

In this research project, the team proved that a novel autonomous radio frequency beam scheduler with optimality guarantees can support secure spectrum sharing of newly available unlicensed and shared 5G millimeter wave bands. This class of distributed schedulers will enable 5G systems to provide additional data throughput and capacity with higher security than currently possible. These improvements will benefit, among others, mission critical communications, such as national security command and control, and emergency response operations in public safety.

The team designed and analyzed the capabilities of a distributed scheduler that considers multiple operators sharing the spectrum in the same geographical area. The co-location of base station antennas was also considered. A game theory approach was used to solve a non-convex sub-problem, and the distributed schedular performance was evaluated. Improvement over existing solutions was demonstrated.

The team developed and analyzed a Q-learning algorithm solution in the reinforcement learning framework. A multiple operators' base station distribution was used to analyze the reinforcement learning effectiveness. The Q-learning algorithm performance was compared to the game theory solution. The team concluded that while both solutions perform satisfactorily with comparable performance in general, the Q-learning outperforms in the interference dominated configurations.



(a) Interference from collocated base station antennas cannot be sensed at the base stations. (b) Results from proposed distributed scheduling that maximizes network utility with spectrum sharing.

TALENT PIPELINE:

- Xiang Zhang, student at University of Utah
- Shamik Sarkar, student at University of Utah

INTELLECTUAL PROPERTY:

Bhuyan, A., M. Ji, and S. Kasera, "Systems, devices, and methods for autonomous beam scheduling for spectrum sharing," PCT U.S. Patent Application 63/107,495 (October 30, 2021).

PUBLICATION:

Zhang, X., S. Sarkar, A. Bhuyan, S. K. Kasera, and M. Ji, "Non-cooperative game-based distributed beam scheduling framework for 5G millimeter-wave cellular networks," *IEEE Transactions on Wireless Communications* 21(1) (2021) 489–504.

PRESENTATIONS:

Zhang, X., S. Sarkar, A. Bhuyan, S. Kasera, and M. Ji, "A Q-learning-based approach for distributed beam scheduling in mmWave networks," *IEEE International Symposium on Dynamic Spectrum Access Networks (DySPAN)*, Dec. 2021.

Zhang, X., S. Sarkar, A. Bhuyan, S. Kasera, and M. Ji, "A non-cooperative game-based approach to distributed beam scheduling in millimeter-wave networks," *Asilomar Conference on Signals, Systems, and Computers, IEEE Signal Processing Societ*y, Nov. 2021.

Zhang, X., S. Sarkar, A. Bhuyan, S. Kasera, and M. Ji, "A stochastic optimization framework for distributed beam scheduling in 5G mm-wave networks over non-cooperative operators," *Asilomar Conference on Signals, Systems, and Computers, IEEE Signal Processing Society*, Nov. 2020.

Sarkar, S., X. Zhang, A. Bhuyan, M. Ji, and S. Kasera, "Enabling uncoordinated spectrum sharing in millimeter wave networks using carrier sensing," *Asilomar Conference on Signals, Systems, and Computers, IEEE Signal Processing Society*, Nov. 2020.

Orthogonal Frequency-division Multiplexing-compatible Underlay Communication Channel for Massive Machine-type Communications in Fifth Generation and Beyond

TOTAL APPROVED AMOUNT:
$789,095 over 3 years

PROJECT NUMBER:
19A39-137

PRINCIPAL INVESTIGATOR:
Arslan Majid

CO-INVESTIGATORS:
Hussein Moradi, INL
Behrouz Farhang-Boroujeny, University of Utah

Advancing technology that enables the Internet of Things applications, such as smart grid, smart home/city, and sensor networks.

Cyclic prefixed direct sequence spread spectrum (CP-DSSS) signaling was used as a data channel for passing a small amount of information between devices in an underlay mode while avoiding any significant interference to mobile broad band communications. CP-DSSS enables a parallel network to the Long Term Evolution (LTE) network for the new massive machine type communication applications. The LTE network and the proposed network of devices will co-exist over the same spectral resources with a minimum amount of interference among them. Compared to competing technologies, CP-DSSS allows for communications in the low signal to noise ratio regime. It can be spectrally isolated from primary users and the waveform can trivially carry out low-complexity multi-user and multi-antenna communications. All these properties are key in establishing CP-DSSS as an underlay femtocell network technology that can co-exist with 5G and beyond primary network communications in both time and frequency.

The CP-DSSS capacity was compared to the ubiquitous orthogonal frequency-division multiplexing technology, where it was shown that CP-DSSS achieves the same capacity as orthogonal frequency-division multiplexing. The unique signaling of CP-DSSS offers a simple symbol rate reduction technique that greatly reduces CP-DSSS transceiver complexity at low signal to noise ratios when compared to orthogonal frequency-division multiplexing. This important finding distinguishes CP-DSSS for low-powered operation. In addition, it establishes CP-DSSS as a suitable underlay technology for femtocell network communications which share the same spectrum as the primary network.

One goal of this research was to identify an optimal or near-optimal detector for the waveform. The usefulness of such a detector allows the waveform to establish links that approach channel capacity. It was found that a combination of match-filter detection and time-reversal precoding are sufficient for symbol detection. A frequency-domain multi-user detector design using a regularized zero-forcing precoder was proposed to further reduce the computational cost in both the uplink and downlink communications. Critical to the design of this precoder was devising a mechanism to reuse the large matrix inverses necessary for carrying out the calculations involved in transceiver precoding and detection.

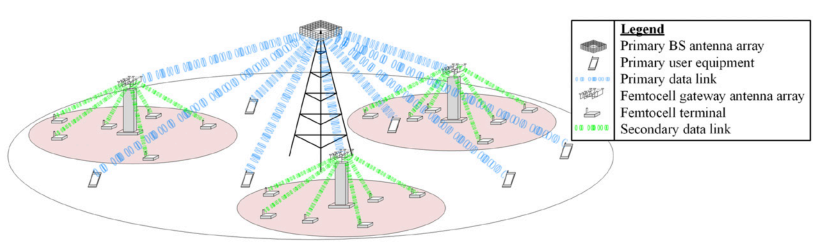


Illustration of a CP-DSSS based femtocell network that synchronizes with 5G and beyond primary base station (BS) networks and establishes a secondary network of CP-DSSS communications.

TALENT PIPELINE:

- Stephen Jenkins, student at University of Utah
- Brent Kenney, student at University of Utah
- Austin Stevens, student at University of Utah
- Dallin Hansen, student at University of Utah

INTELLECTUAL PROPERTY:

Farhang-Boroujeny, B., A. J. Majid, and H. Moradi, "A method of spread spectrum modulation for communications in femtocell networks." U.S. Patent Application No. 17/249,593 (March 9, 2019).

PUBLICATION:

Kenney, B. A., A.J. Majid, H. Moradi, and B. Farhang-Boroujeny, "Frequency domain detection and precoding for massive MIMO with single carrier modulation," *IEEE Transactions on Wireless Communications* (2021) Ahead-of-Print. Available at: www.doi.org/10.1109/TWC.2021.3119486.

PRESENTATIONS:

Kenney, B. A., S. N. Jenkins, A. J. Majid, H. Moradi, and B. Farhang-Boroujeny, "Cyclic prefix direct sequence spread spectrum capacity analysis," *2020 IEEE 92nd Vehicular Technology Conference* (2020-Fall).

Jenkins, S., B. Kenney, A. Majid, H. Moradi, and B. Farhang-Boroujeny, "CP-DSSS — A novel waveform for multiple access in IoT," *IEEE 6G Summit*, Mar. 17, 2020.

Kenney, B. A., A. J. Majid, H. Moradi, and B. Farhang-Boroujeny, "Near optimality of matched filter detection for cyclic prefix direct sequence spread spectrum," *2nd 6G Wireless Summit 2020*, Mar. 17, 2020.

Kenney, B. A., A. J. Majid, H. Moradi, and B. Farhang-Boroujeny, "Multi-user capacity of cyclic prefix direct sequence spread spectrum with linear detection and precoding," *2020 IEEE 92nd Vehicular Technology Conference* (2020-Fall).

Kenney, B. A., A. J. Majid, H. Moradi, and B. Farhang-Boroujeny, "Frequency domain multi-user detection for single carrier modulation with cyclic prefix," *GLOBECOM 2020 - 2020 IEEE Global Communications Conference*.

Farhang-Boroujeny, B., A. J. Majid, and H. Moradi, "CP-DSSS — An OFDM compatible variable rate modulation for 5G and beyond," *2020 IEEE 3rd 5G World Forum* (5GWF).

Kenney, B. A., A. J. Majid, H. Moradi, and B. Farhang-Boroujeny, "Opportunistic secondary communications downlink using multi-antenna femtocell gateways," *WiMob 2021 - Wireless and Mobile Computing, Networking, and Communication*s.

Kenney, B. A., A. J. Majid, H. Moradi, and B. Farhang-Boroujeny, "Efficient precoding for single carrier modulation in multi-user massive MIMO networks," *2021 - IEEE International Conference on Communications*.

Risk and Resilience Assessment of Cyberattacks on Electric Grids: Informing Risk Characterization Using Dynamic Probabilistic Risk Assessment

TOTAL APPROVED AMOUNT:
$1,560,000 over 3 years

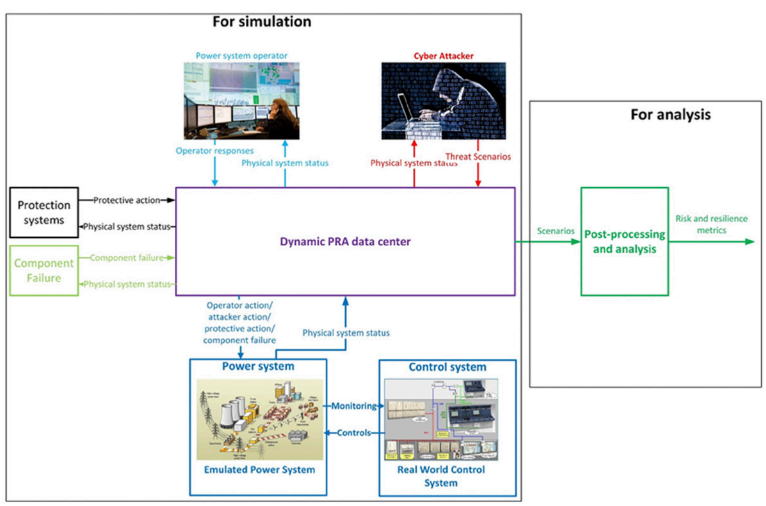
PROJECT NUMBER:
19P45-020

PRINCIPAL INVESTIGATOR:
Katya Le Blanc

CO-INVESTIGATORS:
Craig Rieger, INL
Thomas Ulrich, INL
Timothy McJunkin, INL
Brian Johnson, University of Idaho
Carol Smidts, The Ohio State University

Advancing the scientific understanding needed to develop a methodology for evaluating risk in high-consequence cyber-physical systems that make up critical infrastructure.

Understanding risk in high-consequence cyber-physical systems required characterizing the complex interactions between physical systems and general-purpose computing components that are distributed across those systems. Current approaches rely on subjective assessment based on subject matter expert input and do not have a firm scientific basis. There are mature and accepted methods for understanding risk in physical systems that require a thorough understanding of the components in the system, how those components interact to perform their intended functions, and the likelihood of failure of the components. Cyber-physical systems have another layer of complexity because they can aggregate previously unknown interactions between components in the physical system through intentional or unintentional misuse of the general computing capability of the cyber technologies embedded in the system. When the threat is an intelligent adversary, whose goal is to compromise or sabotage the system, the complexity of determining what can happen and how likely it is becomes ever more difficult. The horizontal application of the same or similar cyber technologies creates a scale in attack surface complexity that can overwhelm the present high-fidelity "n-k" physical failure models with orders of magnitude greater numbers and modalities. More computational power is necessary but not sufficient to provide the solutions that will be needed by investment decision makers. A scientific basis for decision tools and sensitivity analysis is crucial to guide the development and application of higher-fidelity physical effects models to the most consequential assets and to guide critical choices with uncertainty in data-limited scenarios. This project addressed these critical gaps by developing a theoretical framework for characterizing risk and uncertainty, collecting and generating data using modeling and simulation techniques, and developing repeatable methodology that can scale in complexity and application for risk quantification.



Dynamic probabilistic risk assessment framework.

TALENT PIPELINE:

- Katherine Herdt, student at Virginia Polytechnic Institute and State University
- Xiaoxu Diao, postdoc at The Ohio State University

PUBLICATIONS:

Le Blanc, K., "Human factors challenges in developing cyber-informed risk assessment for critical infrastructure," In: *Advances in Artificial Intelligence, Software and Systems Engineering. AHFE 2020. Advances in Intelligent Systems and Computing*, Ahram, T. (ed.), vol 1213. Springer, Cham (2021).

Li, R., and K. Le Blanc, "Approaches to human performance modeling of electric grids operators," In: *Advances in Safety Management and Human Performance. AHFE 2020, Advances in Intelligent Systems and Computing*, Arezes, P., and R. Boring (eds.), vol 1204. Springer, Cham (2021).

Smadi, A., B. T. Ajao, B. K. Johnson, H. Lei, Y. Chakhchoukh, and Q. A. Al-Haija, "A comprehensive survey on cyber-physical smart grid testbed architectures: Requirements and challenges," *Electronics* 10(9) (2021) 1043.

PRESENTATIONS:

Diao, X., Y. Zhao, C. Smidts, H. Lei, and B. K. Johnson, "Dynamic probabilistic risk assessment for electric grid cybersecurity risk assessment." *International Topical Meeting on Probabilistic Safety Assessment and Analysis 2021*.

Gui, J., H. Lei, Y. Chakhchoukh, T. R. McJunkin, and B. K. Johnson, "Transmission adaptive capacity-based resilience metrics for power grid contingency analysis," *53rd IEEE North American Power Symposium*, Nov. 2021.

Lingaraju, K., J. Gui, B. K. Johnson, and Y. Chakhchoukh, "Simulation of the effect of false data injection attacks on supervisory control and data acquisition using PSCAD/EMTDC," *52nd IEEE North American Power Symposium*, Apr. 2021.

Zhao, Y., M. Zhu, and C. Smidts, "Data post-processing for dynamic probabilistic risk assessment," *International Topical Meeting on Probabilistic Safety Assessment and Analysis* (2021).

Li, R., T. R. McJunkin, and K. Le Blanc, "Integrating operator function in work domain analysis of electric grid operations," *Human Factors and Ergonomics Society Annual Meeting* (Dec. 2020), Vol. 64, No. 1, pp. 170307.

Zhao, Y., N. Adina, K. Le Blanc, C. Rieger, B. K. Johnson, H. Lei, T. McJunkin, P. K. Vaddi, T. Ulrich, R. Li, and C. Smidts, "Dynamic probabilistic risk assessment for cyber security risk analysis of the electric grid," *The 30th European Safety and Reliability Conference and the 15th Probabilistic Safety Assessment and Management Conference* (2020).

Le Blanc, K., "Computer security in the energy industry: The security risks of ignoring the human in engineering and design," *Boise State Computing Colloquium* (2020).

Smart Contingency Analysis Neural Network for in-depth Power Grid Vulnerability Analyses

TOTAL APPROVED AMOUNT:
$120,000 over 1 year

PROJECT NUMBER:
20A1047-038

PRINCIPAL INVESTIGATOR:
Bjorn Vaagensmith

CO-INVESTIGATOR:
Sam Yang, Florida State University

Combining fundamental physics-based relationships in the electrical grid infrastructure with artificial intelligence-based analyses enables power utility providers to explore and discover system vulnerabilities before problems occur.

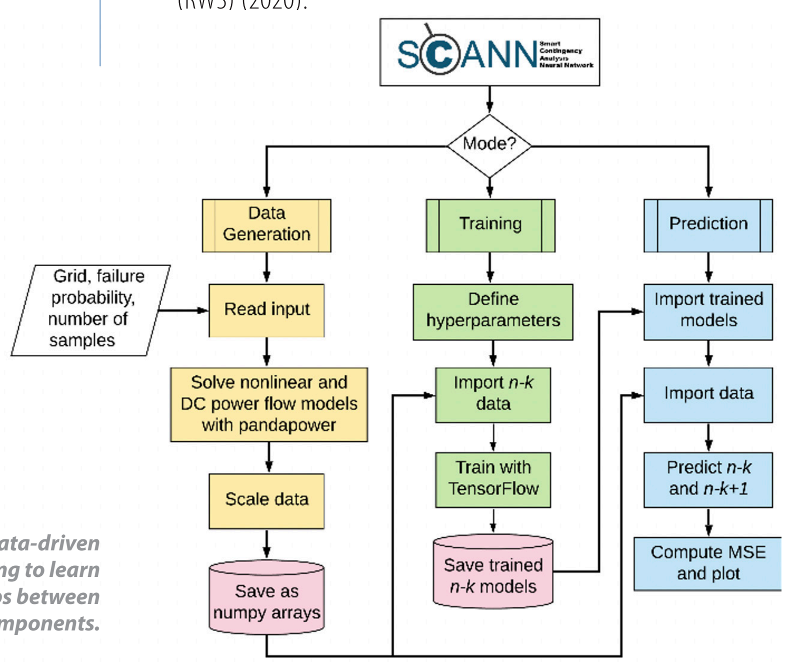
Contingency analyses are crucial to prevent detrimental power outages by enabling utilities to discover vulnerabilities within their grid. An n-k contingency is subject to k number of component failures caused by severe weather conditions, software failure (e.g., 2003 Northeastern blackout), or cyberattacks (e.g., 2019 denial-of-service attack on the Western transmission grid). However, power utilities are typically limited to n-1 system-wide contingency analysis due to the combinatorial nature of the problem—the computational cost required to sweep through all possible power flow simulations increases exponentially as a function of k. This research investigated the potential of a smart contingency analysis neural network (SCANN) framework based on machine learning to reduce the computational expense normally required to calculate all possible n-k contingencies by half. SCANN is used to discover higher order contingencies up to n-3, after which the prediction error becomes greater than the acceptable tolerance. Additionally, SCANN can compute results 147 times faster than the Newton-Raphson solver used in conventional power grid analyses. Based on this learning with SCANN, which uses residual neural networks, a new research focus into the resiliency of the grid using Bayesian neural networks can be explored.

TALENT PIPELINE:

- Deepika Patra, student at Arizona State University.

PRESENTATION:

- Yang, S., B. Vaagensmith, and D. Patra, "Power grid contingency analysis with machine learning: A brief survey and prospects," IEEE Resilience Week (RWS) (2020).



SCANN uses data-driven encoding to learn relationships between power grid components.

Additive Manufacturing Control of Defects

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

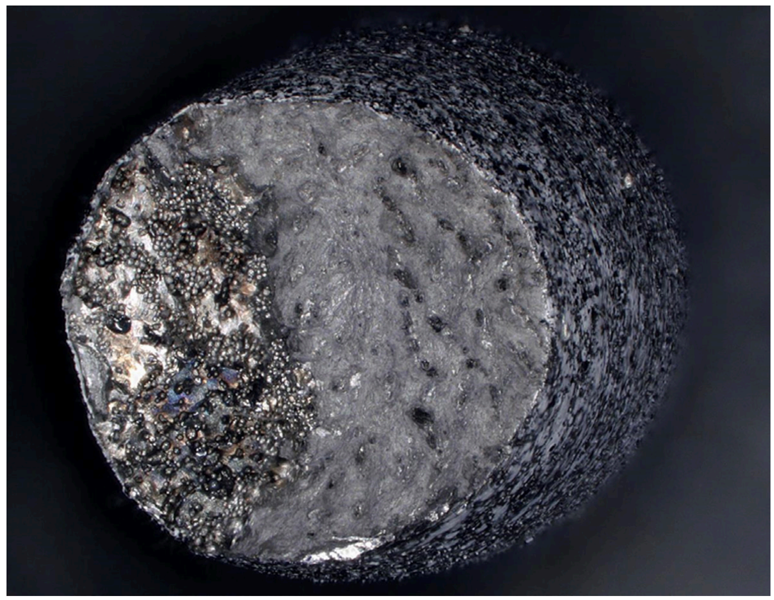
PROJECT NUMBER:
20A1047-048FP

PRINCIPAL INVESTIGATOR:
Natalie Pinkham

CO-INVESTIGATOR:
Thomas Walters, INL

Mechanical properties of additive manufactured parts can be tailored for specific attributes by controlling build parameters.

This project used an Optomec Laser Engineered Net Shaping additive manufacturing system to fabricate parts with repeatable process parameters by employing closed-loop process control. The process control used in situ optical and infrared imaging of the melt pool. Part defects were classified layer-by-layer using computed tomography scanning to correlate a part build. Controlling specific build parameters resulted in desired mechanical properties.



Defect evaluation of an additive manufactured specimen.

Improving Cryptographic Algorithm Resistance to Side Channel Attack

TOTAL APPROVED AMOUNT:
$105,000 over 1 year

PROJECT NUMBER:
20A1052-028

PRINCIPAL INVESTIGATOR:
Robert Erbes

CO-INVESTIGATORS:
Liljana Babinkostova, Boise State University
Mike Borowczak, University of Wyoming

Improving computer system resistance to side channel attack by modifying mathematic primitives used in cipher implementation.

Cryptographic ciphers are commonly used to secure various aspects of computer systems. The security provided by ciphers typically comes from the requirement that a user have knowledge or possession of a key to interact with that system (e.g., reading a message or running a program). Using cryptographic ciphers is particularly important in environments where physical protection of computing equipment is difficult or unfeasible, such as with the Internet of Things or cyber-physical systems. Traditional cryptographic ciphers can be too computationally expensive to run on resource-constrained computing systems, necessitating lightweight ciphers designed to operate within such environments.

However, even ciphers with ideal performance may still be susceptible to attack after their implementation in a multiple of mediums (e.g., processor, computer, electro-mechanical system, or simulator). Observations of the side effects of computation, such as power usage, radio emissions, heat generation, or instruction timing, can be used to infer otherwise secret information about the data being processed. This observation and inference are often referred to as a 'side channel' attack. Cryptographic implementations deployed on resource-constrained devices are especially vulnerable to these types of attacks.

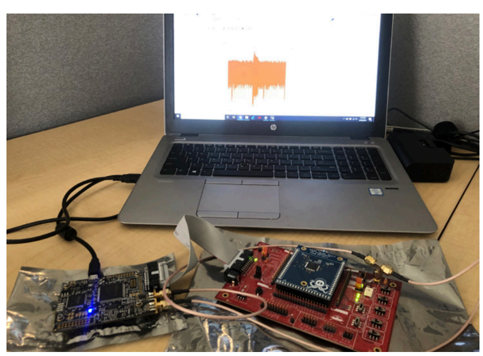
This project tested susceptibility to power-based side-channel attacks on a common platform of several National Institute of Standards and Technology lightweight cipher candidates, evaluated existing published metrics for predicting susceptibility to attack, and implemented candidate ciphers using different algorithmic variants. The team discovered that all but one of the existing published metrics used to evaluate components of substitution-permutation network-based cryptographic ciphers do not predict resistance to side channel attack.

TALENT PIPELINE:

- William Unger, student at Boise State University

PRESENTATION:

Unger, W., L. Babinkostova, M. Borowczak, and R. Erbes, "Side-channel leakage assessment metrics: A case study of GIFT block ciphers," 2021 IEEE Computer Society Annual Symposium on Very Large-Scale Integration (July 2021).



The ChipWhisperer open-source system hardware used in this research (O'Flynn and Chen, International Workshop on Constructive Side-Channel Analysis and Secure Design, 2014). The correlation power analysis attacks and data analysis were performed using Python.

Infrastructure eXpression

TOTAL APPROVED AMOUNT:
$735,000 over 2 years

PROJECT NUMBER:
20A44-025

PRINCIPAL INVESTIGATOR:
Rita Foster

CO-INVESTIGATORS:
Ryan Hruska, INL
John-Mark Gurney, New Context Services, Inc.

Researchers codified an attack surface into an international data standard, enabling new defense modeling for better infrastructure protection.

The goals of this project were to use an international data standard to describe critical infrastructure digital configurations, enable automated cyber detection and response at near machine speeds, and provide context rich infrastructure modeling using graph theoretics. The project produced the software, Infrastructure eXpression, which automatically discovers configuration components and creates full attack surfaces, including the normally hidden components in hardware, networks, and firmware. Infrastructure eXpression implements a common language for systems and their potential cyber issues, which provides the framework for automated discovery, detection, and response.

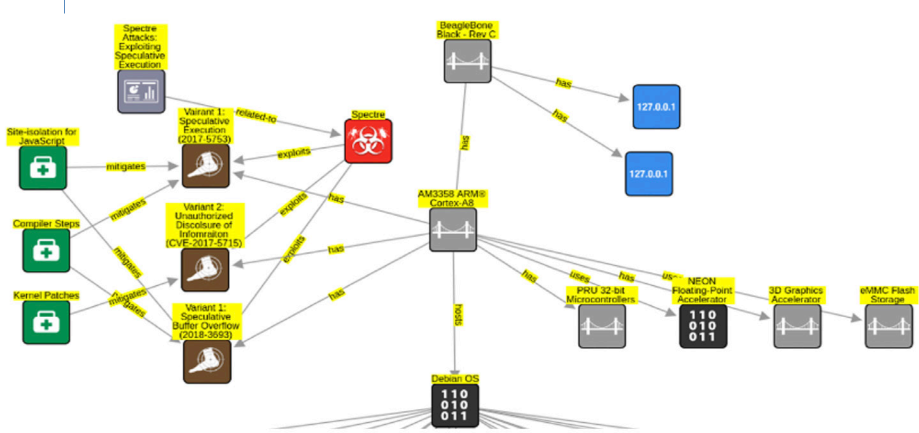
The largest research challenge was to implement a draft standard that has never been used. Many different techniques were attempted to codify the wide variety of infrastructure information needed to enable the advanced data analytics capabilities in graph theoretics. Data science advances in nodes, edges, and relationships enabled the representations of disparate data types (e.g., temporal network session and more static operating systems). After validating the data structures, mathematical measures for similarities to the evidence-based infrastructure observables is possible. The test infrastructure was analyzed for vertex clustering, communities, and cliques within a subgraph. Clique discovery used connected edges for a pair of nodes; other similarities only show connection to one node.

TALENT PIPELINE:

- Isabella Magallanez, student at the University of Texas at San Antonio
- Karina Permann, student at University of Idaho
- Manual Maestas, student at Idaho State University
- Shaya Wolf, student at University of Wyoming

PRESENTATION:

Foster, R., Z. Priest, and M. Cutshaw, "Infrastructure eXpression for codified cyber-attack surfaces and automated applicability," *IEEE Resilience Week* (RWS) (2021).



Partial attack surface showing hardware components, malware, vulnerabilities, and courses of action. (Image generated from the Structured Threat Intelligence Graph).

Automated Type and Data Structure Resolution

TOTAL APPROVED AMOUNT:
$550,000 over 2 years

PROJECT NUMBER:
20A44-108

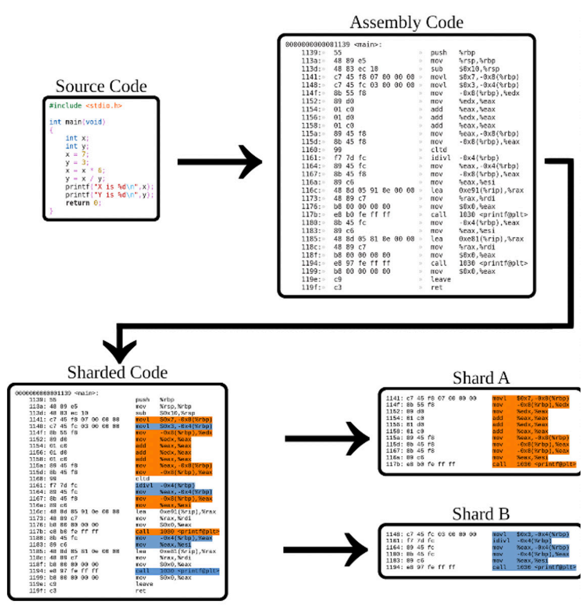
PRINCIPAL INVESTIGATOR:
Jared Verba

CO-INVESTIGATOR:
Sean Salinas, INL

Discerning data type information from compiled binaries using machine learning has the potential to enable better preparation against and response to malware attacks.

Research was conducted to enhance the ability for reverse engineers to take compiled binaries and disassemble them to gain more information about their inner workings. This project specifically explored pathways to automatically discern data structure and data type information from compiled binaries, primarily using machine learning techniques. An additional benefit of automating the process would allow for greater scale of binaries to be reverse engineered without manual type resolution. The research: (1) developed a training data set using compiled binaries and known type information; (2) expanded the training set so that the machine learning algorithms could recognize a diverse set of type information from the compiled binaries; (3) experimented with machine learning processes against an unknown binary (one in which the source code is not available) so that we could automatically discern type information from it without user intervention; and (4) analyzed how this newly discerned type information might enable reverse-engineers to focus on other interesting parts of the compiled code instead of manually resolving data types.

Research results indicated that discerning type information using machine learning algorithms was very sensitive to conditions and may only be applicable to specialized environments. The algorithms rarely gave good results. When the results were good, they were specific to the data set that was used for training. When properly trained and tuned, the algorithms would give more positive results on problems similar to their training set. However, when extrapolated outside of its training set norms, the algorithms would give poor results that required user verification and manual intervention. Further refinement of the solution is required before meaningful application of this technique can be developed.



Example of the sharding process that simulates every section of memory that the binary references and keeps track of what machine instructions reference each section. Using this technique, large instruction sets were generated for each memory reference and were then used for machine learning.

Support Vector Analysis for Computational Risk Assessment, Decision-Making, and Vulnerability Discovery in Complex Systems

TOTAL APPROVED AMOUNT:
$125,000 over 1 year

PROJECT NUMBER:
20A1054-008

PRINCIPAL INVESTIGATOR:
Andrei Gribok

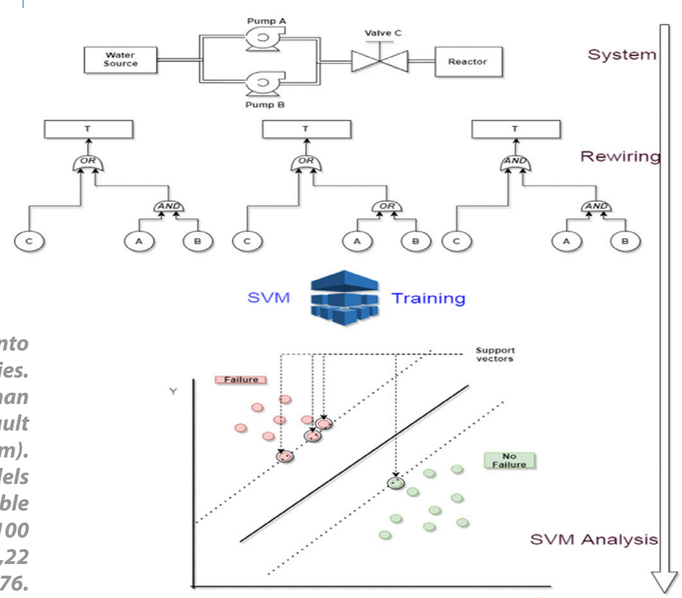
CO-INVESTIGATOR:
Curtis Smith, INL

This research improves risk quantification, reduces human error in risk assessment, and enables subject matter experts to focus on critical risk areas.

Modern probabilistic risk assessment is a method of documenting the discoveries of an analyst based on their qualifications and available system information. This is not a process to suggest new, previously unknown risk. The purpose of this research was to develop methods of auto-detecting possible vulnerabilities in system designs to uncover previously unseen issues and reduce human error/costs by allowing analysts to focus on critical areas via intelligent, efficient sampling of the system's parameter space.

The objective was to develop a fundamentally new way of exploring supervised binary classification using support vector machines to intelligently guide the sampling process through very high-dimensional parameter spaces to analyze logical flaws in the system's complex design. Currently, the machine learning field is dominated by pattern recognition, data representation, and forecasting. Research in machine learning techniques to discover logical fallacies is lacking. The newly developed methodology uses machine learning to develop a computational method for analyzing logical constructs (e.g., fault trees) represented as Boolean expressions.

This project produced two primary outcomes. This first is a new, broadly applicable methodology that uses intelligently guided space sampling methods to drastically reduce the number of system configurations needing to be analyzed. This methodology allows researchers to auto-detect possible vulnerabilities in system designs, devices, and networks to uncover previously unseen issues and reduce human error and costs by enabling analysts to focus on critical areas via intelligent, efficient sampling of the system's parameter space. The second outcome is a demonstration of the benefits of machine learning, as applied to a case study relating a computational risk assessment applied to probabilistic risk assessment.



Risk assessment gives insights into potential system vulnerabilities. Historically modeled by human experts and represented via fault trees (logic model for the system). However, the process and models are complex. The number of possible paths to failure for a system with 100 components is 1,267,650,600,228,229,401,496,703,205,376.

Application of the Associated Particle Technique to Fieldable Prompt Gamma-Ray Neutron Activation Analysis Systems

TOTAL APPROVED AMOUNT:
$80,000 over 1 year

PROJECT NUMBER:
21A1055-016

PRINCIPAL INVESTIGATOR:
Brian Bucher

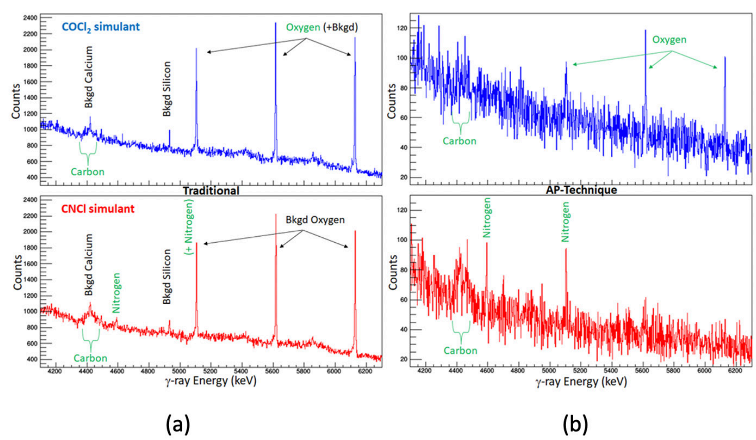
CO-INVESTIGATOR:
Jayson Wharton, INL

Improving the sensitivity and speed of nondestructive detection of chemical and biological agents in hidden threat objects.

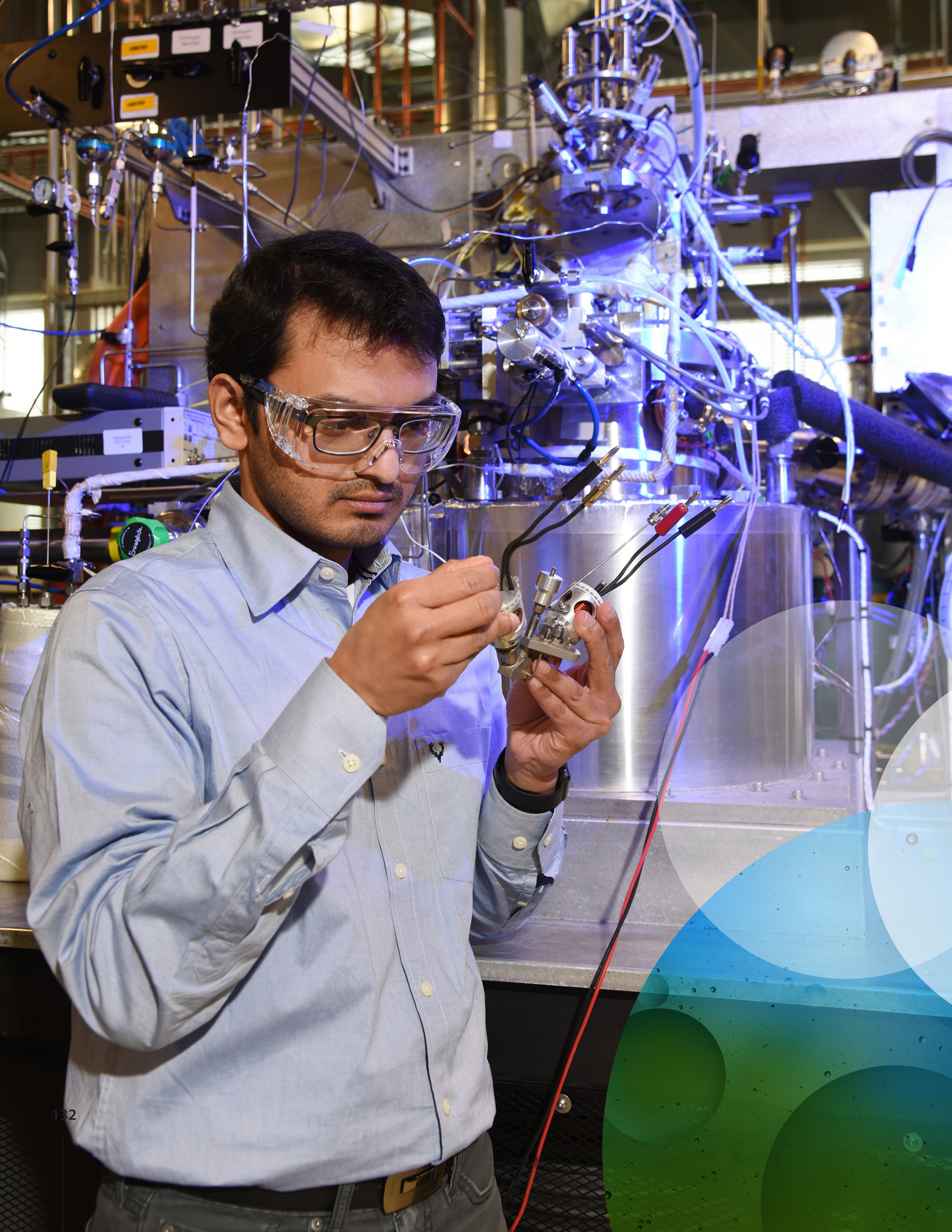
Prompt gamma-ray neutron activation analysis is useful for determining the chemical elements inside an unknown object. This is done using a source of neutrons to irradiate the object, which induces characteristic gamma rays through nuclear reactions with the constituent atoms. The gamma rays are then detected and identified using an external spectrometer. Deuterium-tritium neutron generators are useful because they produce energetic 14 MeV neutrons that are highly penetrating and able to excite nuclei within light atoms that typically require a high threshold energy to induce gamma-ray emission. However, these energetic neutrons easily excite nuclei in the surrounding environment, which causes a large background signal in the gamma-ray spectrometer. This study investigated the potential of the associated particle technique to suppress the high background signal. The associated particle technique uses a specialized neutron generator that can identify the precise moment in time when a neutron is emitted in the direction of the test object, thus providing a gate signal for the spectrometer to identify which detected gamma rays are coincident with neutrons that were directed toward the sample and which gamma rays likely came from the surrounding environment.

The tested samples were field assays of armaments containing chemical warfare agents and high explosives. Three different gamma-ray detectors were examined: sodium iodide, lanthanum bromide, and high-purity germanium. In each case, using the associated particle technique clearly reduced the background. The high-purity germanium was the most promising for field assessments because of its superior energy resolution, despite its much poorer timing resolution compared to the other detectors. Even with the poor gamma-ray time resolution, the associated particle technique suppressed background radiation to such a low level that a spectral background subtraction was no longer needed.

A representative example relevant to chemical weapons detection is the discrimination of phosgene ($COCl_2$) from cyanogen chloride (CNCl). With traditional prompt gamma-ray neutron activation analysis, these two are nearly indistinguishable due to background from the detected oxygen. Moreover, the intense oxygen lines in the gamma spectra interfere with the strongest nitrogen line, which is usually quite weak, making the latter very difficult to identify. The associated particle technique eliminated oxygen from the CNCl spectrum, which made the nitrogen signal clearly visible. Furthermore, the low oxygen signal from $COCl_2$ remained in its respective spectrum with all the background eliminated. Therefore, the distinction was clear by analyzing the two spectra with no background subtraction or quantitative analysis needed and without any additional run time beyond the standard 1-hour assays typically employed in these scenarios.



Measurements of simulated $COCl_2$ (top) and CNCl (bottom) in 4.2 in. mortar projectiles. (a) The spectra obtained from traditional prompt gamma-ray neutron activation analysis look nearly identical. (b) The spectra obtained using the associated particle method eliminates the background oxygen making the two chemicals easily distinguishable.



"Everything is theoretically impossible, until it is done. One could write a history of science in reverse by assembling the solemn pronouncements of highest authority about what could not be done and could never happen."

— Between Planets, Robert A. Heinlein, Science Fiction Author

Idaho National Laboratory