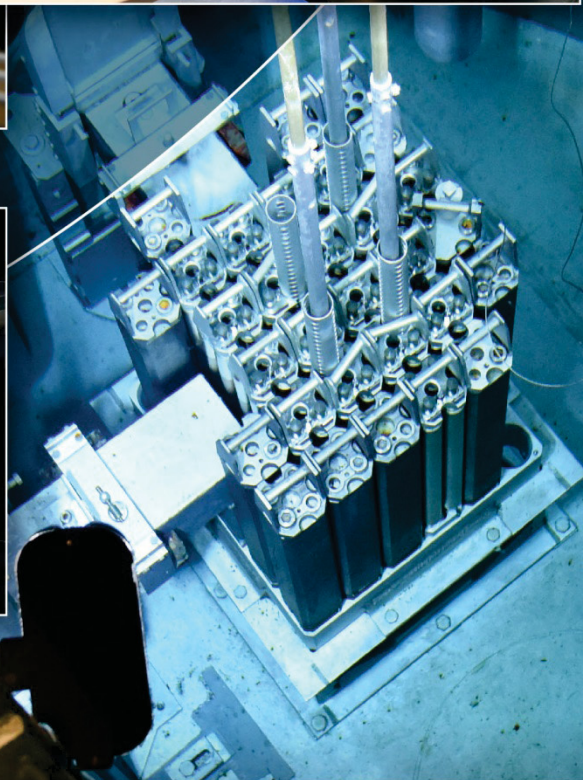
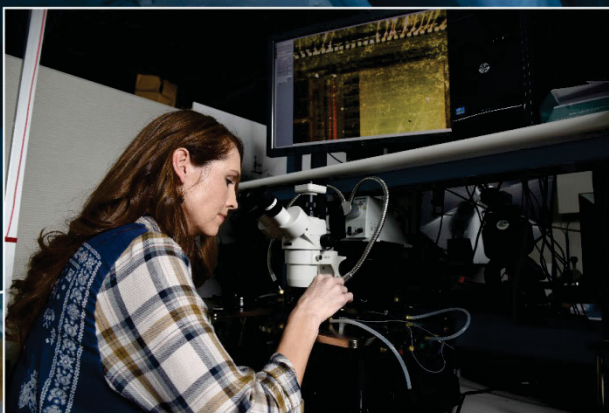


Laboratory Directed Research and Development

# 2016 ANNUAL SUMMARY REPORT

January 2017



On the cover:

**Mr. Troy Unruh, Nuclear Instrumentation Researcher:** Mr. Troy Unruh's research focuses on developing advanced in-core instrumentation for irradiation testing experiments. This research leads to a real-time understanding of the behavior of new and innovative nuclear reactor fuels and materials under development for both the current fleet of nuclear reactors and the next generation of nuclear reactors.

**Dr. Rebecca Fushimi:** Dr. Rebecca Fushimi works in the area of interfacial chemistry and catalysis. Her background includes selective oxidation, dehydrogenation, and reforming reactions on supported metals and mixed metal oxide catalysts. She has been working with the temporal analysis of products transient kinetic technique for more than 15 years and is the leading expert in the temporal analysis of products experimental methodology. A key focus of her work is to apply this unique tool to accelerate industrial catalyst development.

**Ms. May Chaffin, Computer Security Researcher:** May Chaffin's research focuses on control systems cybersecurity that seeks to mitigate cyber threats and vulnerabilities found in industrial control systems and enhance the resilience of operational systems. This research increases knowledge and spearheads groundbreaking technologies designed to incorporate resilience into critical control system components.

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## ACRONYMS

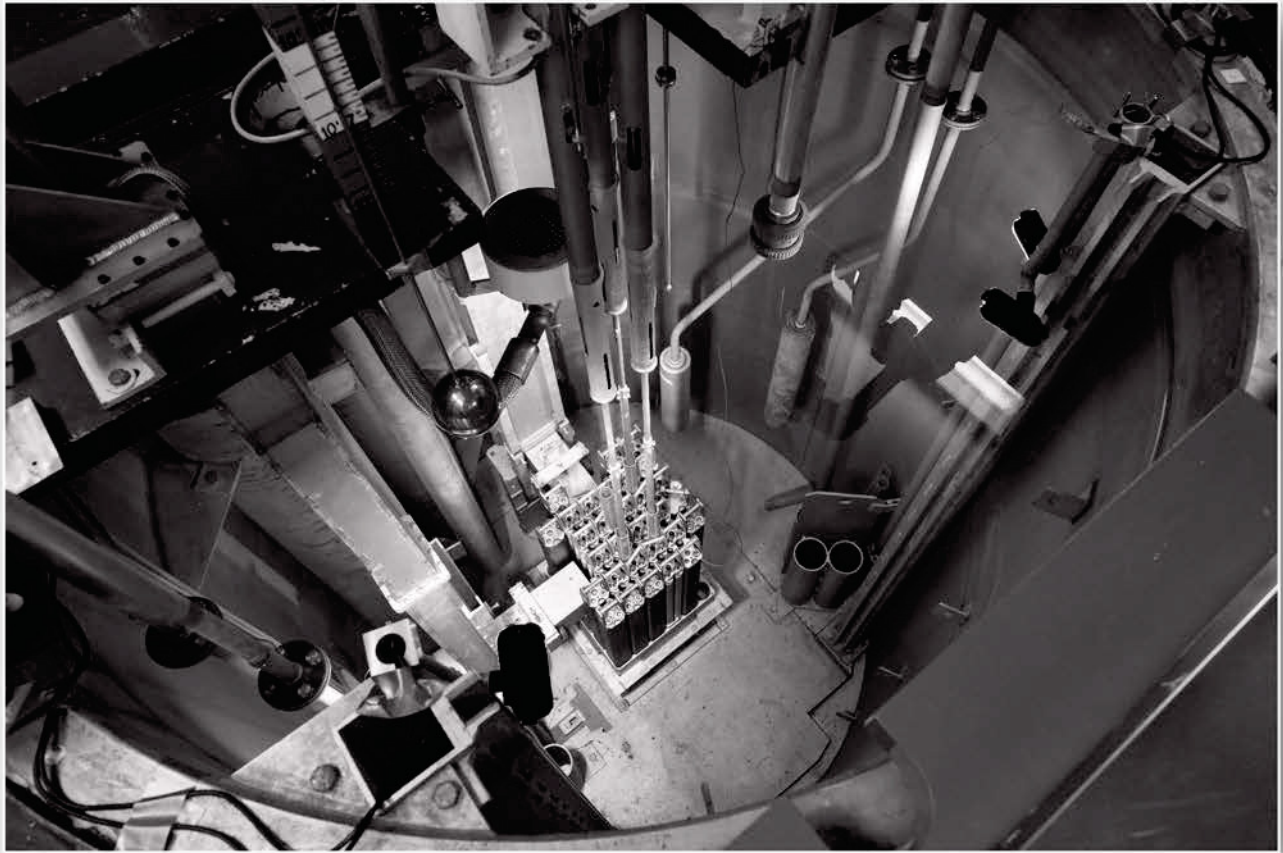
3SBD	safety, security, and safeguards by design
AEM	advanced electrolyte model
AKUFVE	apparatus for continuous measurement of distribution factors in solvent extraction (translation of Swedish abbreviation to English)
AMI	acoustic measurement infrastructure
AMS	accelerator mass spectrometer
ANE	advanced nuclear energy
ANI	active neutron interrogation
ARTIST	Advanced Reactor Technology Integral System Test
ASTM	American Society of Testing Materials
ATF	accident-tolerant fuel
ATR	Advanced Test Reactor
CAES	Center for Advanced Energy Studies
CAN	controller area network
CCE	consequence-driven, cyber-informed engineering
CD	cluster dynamics
CDS	change detection systems
CEA	French Alternative Energies and Atomic Energy Commission
CED	clean energy deployment
CFD	computational fluid dynamics
CI	critical infrastructure
CIP	critical infrastructure
CIVET	continuous integration, verification, enhancement, and testing
CNT	carbon nanotube
CO <sub>2</sub>	carbon dioxide
CZM	cohesive zone model
DCFC	direct current fast charger
DER	distributed energy resource
DFM	drift-flux model
DFT	density functional theory
DOE	U.S. Department of Energy
DOE-NE	U.S. Department of Energy Office of Nuclear Energy

DTF	density functional theory or distributed test facility
DTPA	diethylenetriaminepentaacetic acid
EMImFHF	1-ethyl-3-methylimidazolium fluorohydrogenate
ESS	energy storage system
EXAFS	extended x-ray fine structure
FIB	focused-ion beam
FO	forward osmosis
FY	fiscal year
GB	grain boundary
HES	hybrid energy system
HEU	highly enriched uranium
HIL	hardware in the loop
ICS	industrial control system
IHTL	intermediate heat transport loop
IL	ionic liquids
INL	Idaho National Laboratory
IPWR	integral pressurized water reactor
IR	infrared
IRMPD	infrared multi-photon dissociation
L/D	length-to-diameter
LDRD	laboratory-directed research and development
LIB	Li-ion batteries
LN	lanthanide
LOCA	loss-of-coolant accident
LVDT	linear variable differential transformer
LWR	light-water reactor
MA	minor actinide
MASLWR	multi-application light water reactor
MCP	micro-channel plate
MIT	Massachusetts Institute of Technology
MOOSE	Multi-Physics Object-Oriented Simulation Environment
MTR	material and test reactor
NCSU	North Carolina State University



NGAS	next generation armor steel
NRAD	neutron radiography reactor
NREL	National Renewable Energy Laboratory
NUC	National University Consortium
OBR	optical backscatter reflectometer
ODH	oxidative dehydrogenation
ODS	oxide dispersion-strengthened
OSU	Ohio State University
PA	phosphoranimine
PB-FHR	pebble-bed, fluoride-salt-cooled, high-temperature reactor
PCHE	printed circuit heat exchanger
PCP	primary coolant pumps
PLC	programmable logic controller
PRA	probabilistic risk analysis
QD	quantum dots
R&D	research and development
rDG	reconstructed discontinuous Galerkin
ROM	reduced order models
RTDS	Real-Time Digital Simulator
RTE	radiation transport equation
RTS	real-time simulation
RTPS	real-time process simulator
SEM	scanning electron microscopy
SHADE	scintillation hydro-gel for antineutrino detection
SHINE	scintillation hydro-gel for isotopic neutron emitters
SME	subject matter expert
SNM	special nuclear material
SPP	Strategic Partnership Projects
SPS	spark plasma sintering or switchable polarity solvent
TAP	temporal analysis of product
TEM	transmission electron microscopy
TREAT	Transient Reactor Test
UN	uranium mononitride

UNM	University of New Mexico
UT	ultrasonic thermometers
WiFIRE	wireless radiofrequency signal identification and protocol reverse engineering
XFEM	extended finite element method



# Introduction



## INTRODUCTION

The Laboratory-Directed Research and Development (LDRD) Program at Idaho National Laboratory (INL) reports its status to the U.S. Department of Energy (DOE) by March of each year. The program operates under the authority of DOE Order 413.2C, “Laboratory Directed Research and Development” (April 19, 2006), which establishes DOE’s requirements for the program while providing the laboratory director broad flexibility for program implementation. LDRD funds are obtained through a charge to all INL programs. This report includes summaries of all INL LDRD research activities supported during Fiscal Year (FY) 2016.

INL is the lead laboratory for the DOE Office of Nuclear Energy (DOE-NE). The INL mission is to discover, demonstrate, and secure innovative nuclear energy solutions, other clean energy options, and critical infrastructure with a vision to change the world’s energy future and secure our critical infrastructure. Operating since 1949, INL is the nation’s leading research, development, and demonstration center for nuclear energy, including nuclear nonproliferation and physical and cyber-based protection of energy systems and critical infrastructure, as well as integrated energy systems research, development, demonstration, and deployment. INL has been managed and operated by Battelle Energy Alliance, LLC (a wholly owned company of Battelle) for DOE since 2005. Battelle Energy Alliance, LLC, is a partnership between Battelle, BWX Technologies, Inc., AECOM, the Electric Power Research Institute, the National University Consortium (Massachusetts Institute of Technology, Ohio State University, North Carolina State University, University of New Mexico, and Oregon State University), and the Idaho university collaborators (i.e., University of Idaho, Idaho State University, and Boise State University).

Since its creation, INL’s research and development (R&D) portfolio has broadened with targeted programs supporting national missions to advance nuclear energy, enable clean energy deployment, and secure and modernize critical infrastructure. INL’s research, development, and demonstration capabilities, its resources, and its unique geography enable integration of scientific discovery, innovation, engineering, operations, and controls into complex large-scale testbeds for discovery, innovation, and demonstration of transformational clean energy and security concepts. These attributes strengthen INL’s leadership as a demonstration laboratory. As a national resource, INL also applies its capabilities and skills to the specific needs of other federal agencies and customers through DOE’s Strategic Partnership Program.

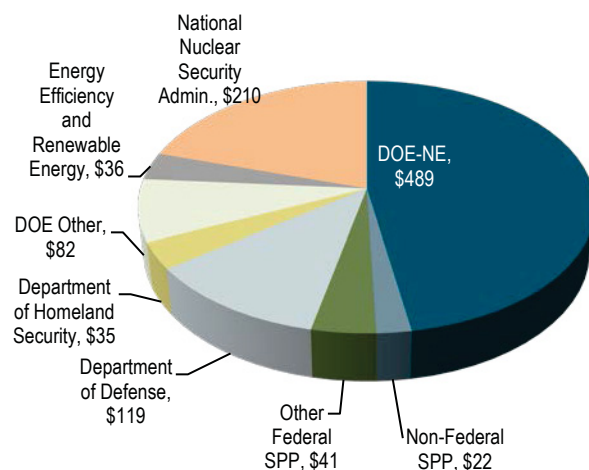
LDRD is a relatively small but vital DOE program that allows INL and other DOE laboratories to select a limited number of R&D projects for the following purposes:

- Maintaining the scientific and technical vitality of INL
- Enhancing INL’s ability to address future DOE missions
- Fostering creativity and stimulating exploration of forefront science and technology
- Serving as a proving ground for new research
- Supporting high-risk, potentially high-value R&D.

Through LDRD, INL is able to improve and accelerate its mission critical outcomes, build/advance its core capabilities, and enhance its ability to conduct cutting-edge R&D for its DOE/National Nuclear Security Administration and Strategic Partnership Program sponsors.

## INL LDRD AT A GLANCE

**Location:** Idaho Falls, Idaho  
**Type:** Multi-Program Laboratory  
**Contractor:** Battelle Energy Alliance, LLC  
**Lead Program Secretarial Office:** DOE-NE  
**Responsible DOE Site Office:** DOE Idaho Operations Office  
**Website:** <http://www.inl.gov/>

**FY 2016 Total Laboratory Operating Costs (excluding Recovery Act): \$1,034M**


**FY 2016 LDRD Program Cost**  
**FY 2016 Funding Cap:** \$30M

**FY 2016 LDRD Investments: 1.9% of Business Volume**

	Projects		Funding (total cost)		Total	
	Continue	New	Continue	New	Projects	Funding
ANE	22	22	\$5,886,019	\$5,109,994	44	\$10,996,014
CED	11	4	\$3,161,781	\$1,059,345	15	\$4,221,130
CIP	7	5	\$1,392,006	\$706,126	12	\$2,098,132
<b>Total</b>	<b>40</b>	<b>31</b>	<b>\$10,439,809</b>	<b>\$6,875,466</b>	<b>71</b>	<b>\$17,313,311</b>

ANE = advanced nuclear energy CED = clean energy deployment CIP = critical infrastructure

**FY 2015 LDRD Program Cost**  
**FY 2015 Funding Cap:** \$28M

**FY15 LDRD Investment: 2.0% of Business Volume**

	Projects		Funding		Total	
	Continue	New	Continue	New	Projects	Funding
ANE	30	17	\$7,666,500	\$3,167,400	47	\$10,833,900
CED	12	6	\$2,874,800	\$1,683,800	18	\$4,558,600
CIP	7	9	\$1,122,800	\$1,266,100	16	\$2,388,900
<b>Total</b>	<b>49</b>	<b>32</b>	<b>\$11,664,100</b>	<b>\$6,117,300</b>	<b>81</b>	<b>\$17,781,400</b>

ANE = advanced nuclear energy CED = clean energy deployment CIP = critical infrastructure

**FY 2015 LDRD Program Cost**  
**FY 2014 Funding Cap:** \$28M

**FY14 LDRD Investment: 2.1% of Business Volume**

	Projects		Funding		Total	
	Continue	New	Continue	New	Projects	Funding
ANE	26	16	\$5,848,900	\$4,113,800	42	\$9,962,700
CED	8	7	\$2,236,900	\$2,032,700	15	\$4,269,600
CIP	4	7	\$933,100	\$1,774,300	11	\$2,707,400
<b>Total</b>	<b>38</b>	<b>30</b>	<b>\$9,018,900.00</b>	<b>\$7,920,800</b>	<b>68</b>	<b>\$16,939,700</b>

ANE = advanced nuclear energy CED = clean energy deployment CIP = critical infrastructure

## LDRD SELECTION PROCESS

Each year, projects are selected for inclusion in the LDRD Program through a proposal process. To select the best and most strategic ideas submitted, the associate laboratory directors responsible for the various mission areas establish committees for the focus area and for transformational funds to review new proposals and ongoing projects. The committees are staffed by senior research and technical managers who are subject matter experts and have no conflict of interest regarding the proposed projects.

Project funding proposals for the Strategic Initiatives R&D Fund, the Transformational R&D Fund, and University Partnership Fund undergo two rounds of review. During the first round, the committees evaluate short preliminary proposals and select the most promising for development into full proposals. During the second round, the committees review the full proposals and ongoing projects that are requesting second or third-year funding. After the reviews are completed, the committees provide funding recommendations to the associate laboratory directors, who, in turn, present the recommendations and their input to the deputy laboratory director for science and technology. The deputy laboratory director develops an overall funding strategy and provides approvals for the investments. All projects selected for funding must also receive concurrence from the DOE Idaho Operations Office.

## LDRD Investment Focus Areas

During FY 2016, \$17.31M was allocated to the INL LDRD Program (Figure 1). This funding supported 71 projects, 31 of which were new starts (Figure 2).

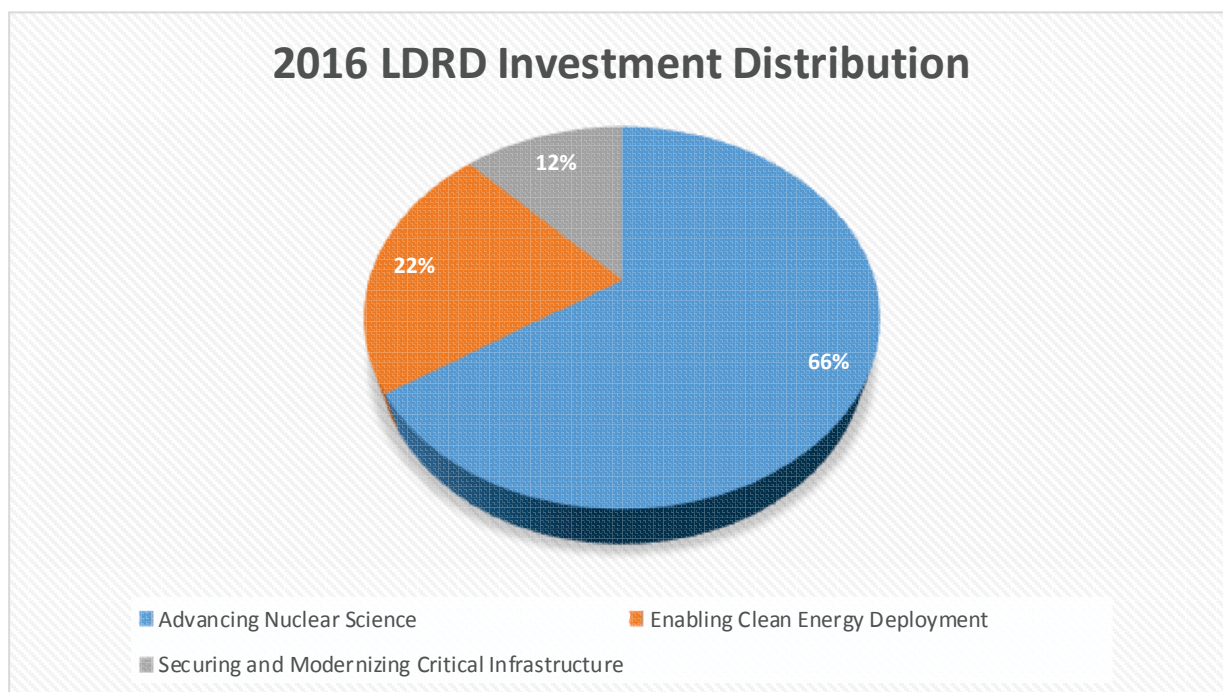
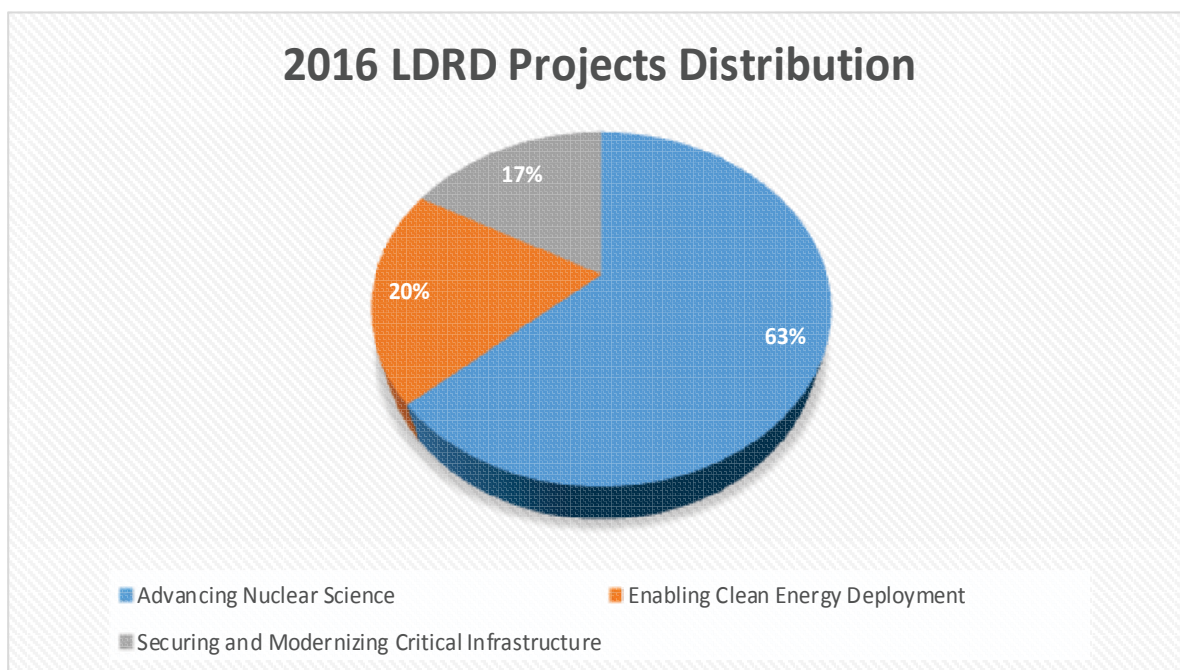


Figure 1. Level of LDRD fund investment by INL mission focus area for FY 2016.





*Figure 2. LDRD projects by INL mission areas for FY 2016.*

To meet the LDRD objectives and fulfill the particular needs of INL, a Strategic Initiatives R&D Fund, investigator-driven Transformational R&D Fund, and University Partnership Fund were established (Table 1). The Strategic Initiatives R&D Fund helps develop new capabilities in support of INL strategic objectives. Transformational Fund Program investments complement the strategic R&D investments by providing a funding source for innovative ideas with potential to enhance INL's core scientific and technical competencies. The University Partnership Fund helps build and sustain university collaboration.

The Strategic Initiatives R&D Fund is the strategic component of the INL LDRD Program and the key tool for addressing the R&D needs of the INL mission priority areas. This transformational program investment supports exploratory research proposals and encourages risk. This program provides a path for funding new approaches that fall within the core capabilities of INL, but outside the targeted research priorities identified in the strategic initiatives. The program may also provide means for addressing a specific technical question of mission interest that will assist in achieving the strategic end states of INL. The University Partnership R&D Fund is the key tool for building and promoting university collaboration and support of R&D needs in the INL mission priority areas. The initiatives, which are the focus of the INL strategy, are the critical areas that INL must concentrate on to be prepared to meet future DOE and national requirements for nuclear energy.

*Table 1. Overview of the strategic initiative R&D, transformational, and university partnership funds.*

	<b>Strategic Initiatives R&amp;D Fund</b>	<b>Transformational Fund</b>	<b>University Partnership Fund (NUC, CAES)</b>
Purpose	Address research priorities of the INL mission areas	Investigator-driven ideas aligned with INL mission areas	Help build university collaboration while addressing research priorities of INL
Reviewers	Mission directorate review committees composed of senior technical managers and subject matter experts	Cross laboratory-wide review committees composed of senior technical managers and subject matter experts	Partnership-led review committees composed of senior technical managers and subject matter experts
Review Process	Preliminary and full proposal review, which may include a presentation to the review committee	Preliminary and full proposal review, which may include a presentation to the review committee	Preliminary and full proposal review, including a presentation to the review committee
Review Cycle	Annual	Annual	Annual
Average Project Budget	Typically about \$240 K/year	Typically about \$50 K/year	Typically about \$224 K/year
Project Duration	12 to 36 months	About 12 months	12 to 36 months
LDRD Outlay	\$15.60M (90% of program)	\$100K (less than 1% of the program)	\$2.02M (10% of the program)

CAES = Center for Advanced Energy Studies  
NUC = National Universities Consortium

## FY 2016 Call Focus Areas

Some of the strategy's success depends to a large extent on INL's ability to identify and nurture cutting-edge science and technology on which enduring capabilities can be built; these are called focus areas. INL uses the resources of the Strategic Initiatives R&D and University Partnership funds to encourage research staff to submit proposals aimed at addressing mission focus area research goals. Each spring, the mission directors issue calls for proposals (i.e., one call for each mission area and one call each for building an NUC and CAES partnership). The call emphasizes specific research priorities that are critical to accomplishing INL's strategic mission areas (those described below are FY 2016 priorities). For the sake of simplicity in this report, each LDRD project has been assigned to a single mission focus area; however, most LDRD projects have applicability to develop core capabilities and unique scientific results that support multiple mission areas (see the appendix to this report for project relevance to various agencies and DOE offices).

### Advancing Nuclear Energy

During FY 2016, INL invested \$10.99M in 44 LDRD projects in support of Advancing Nuclear Energy mission areas covering the following topics:

- **Ensuring long-term reliability of light water reactors and spent fuel storage**, with main focus areas as follows:
  - Advancing nuclear science user facilities capabilities: the objective of this investment is development of advanced instrumentation for both in and out-of-reactor fuels and materials performance, as well as innovative post-irradiation examination capabilities that address challenges associated with testing of large numbers of highly irradiated samples.
  - Fuels and materials R&D modeling and simulation: the objective of this investment is to advance basic development in the areas of (a) advanced fuel fabrication techniques for metal, ceramic, and composite fuels; (b) advanced measurement techniques to characterize fresh and irradiated fuels at the microstructural level; (c) online instrumentation for in-pile experiments; (d) innovative ideas for out-of-pile testing of fuels and materials for targeted phenomenology; (e) innovative ideas for analyzing post-irradiation examination data; and (f) innovations in coated-particle fuel. More specifically, the investments focused on research that coupled experimental studies with multi-scale, multi-physics modeling methods.
  - Transient testing capabilities: the objective of this investment is to create capability for testing fuels during accident conditions such as large power increases and loss-of-cooling events that will provide the safety basis for advanced reactors and fuels. The capability will also provide data that can be used for development, verification, and validation of modeling and simulation tools for fuel system design and development. Focus of the FY 2016 investments is on developing (a) a next generation fuel motion monitoring systems for use during transient testing, (b) novel instruments for supporting separate effects testing (e.g., transient pressure transducers, fast response temperature indicators, and acoustic sensors), and (c) coupled thermal-nuclear reactor performance codes to help design complex transient experiments.
  - Fuel cycle material recovery: the objective of this investment is development of new materials recovery insights ranging from developing fundamental understanding, building a pipeline of experts through postdocs, and developing modeling and simulation tools. The goal is to establish research and process demonstrations with highly radioactive materials.
  - Rapid translation of innovation to the nuclear industry through advances in methods, tools, and concepts for use of existing INL reactors and other experimental facilities. The goal is to increase the use and user base of INL's reactors and major experimental facilities, including the Neutron Radiography Reactor (NRAD), Transient Reactor Test (TREAT), Advanced Test Reactor (ATR), ATR Critical, and Safety and Tritium Applied Research Facility.
- **Facilitate deployment of nuclear systems in the modern energy context**, with main focus areas as follows:
  - Advanced reactor designs and new safety paradigms: the goal of this investment is innovation in advanced reactor designs, including small modular reactor concepts for improving the viability of nuclear power plants and reducing investment risks. Proposals were sought on new reactor concepts, as well as enhancements to existing concepts and designs, adaptation to grid dynamics, load following, hybrid energy systems, and new approaches to dealing with the various sources of investment risk and other potential risk-reducing strategies.
  - Methodologies and tools for validation: the goal of this investment is to create capabilities needed to support challenges such as TREAT restart and analysis of storage and safe management of used nuclear fuel. Specific investment focus is on development of methodologies and tools that would facilitate verification and validation, uncertainty quantification, and scaling analyses through data collection and qualification; creation of a framework to extract information from present data; developing a framework for validation of INL tools; performing uncertainty quantification and data analysis that specifically focus

on experimental data for validation of INL tools; and performing uncertainty quantification and data assimilation using experimental databases.

- Peaceful and secure use of nuclear technology – nuclear nonproliferation: the objective of this investment is to focus on building INL's ability to forecast potential nuclear proliferation risks to help propose and implement U.S. programs that can deter further nuclear weapons development. The emphasis of this research is on (a) safeguards/nonproliferation/forensics-focused nondestructive assay measurements of irradiated fuels within the nuclear fuel cycle; (b) evaluation and demonstration of the integration of safeguards, safety, and security concepts; (c) advancement of capabilities at the Nuclear and Radiological Activity Center to detect and characterize special nuclear materials.

### Enabling Clean Energy Deployment

During FY 2016, INL invested \$4.22M in 15 LDRD projects in support of Enabling Clean Energy Deployment mission areas. The objective of this investment is to advance research in (a) clean energy generation and integration through deployment and integration of renewable energy supply technologies, including energy systems integration, dynamic modeling, simulation, design, test, control, and validation; (b) accelerate deployment of next generation transportation systems, including science-based performance assessments for advancing energy storage, improving performance and affordability of electric vehicles, and advancing bioenergy and biofuels technologies; and (c) advanced manufacturing through advances in sustainable and efficient manufacturing for the United States. Focus of this investment will be on areas in (a) recycling, reuse, energy-water optimization, and net-zero manufacturing; (b) separation of rare-earth elements in manufacturing; (c) development of advanced monitoring, controls, and analysis techniques to improve manufacturing quality; and (d) performance assessment of materials in harsh environments.

### Securing and Modernizing Critical Infrastructure

During FY 2016, INL invested \$2.10M in 12 LDRD projects in support of DOE's Securing and Modernizing Critical Infrastructure mission areas, which included the following:

- Critical infrastructure protection at the nexus of controls, cyber, and wireless: the objective of this investment is to help advance INL's unique and differentiating capabilities in the technology focus areas of cybersecurity, industrial control system security, wireless communications spectrum sharing, power grid security, and material technologies. The investments focused on critical infrastructure protection cybersecurity research that emphasized science-based innovative approaches for inter and intra-system communication that assesses critical system and/or component vulnerabilities. They also focused on the homeland security investment objective to create advances in the areas of (a) resilience of industrial control systems, (b) integrated cyber/physical analysis, (c) assurance of public safety communications, (d) assessment of infrastructure vulnerabilities, dependencies/interdependencies, and resilience, and (e) protection of infrastructure from an all hazards threat. Investment in Wireless National User Facility projects are expected to advance testbed capabilities to conduct tests for commercial, federal/government, and academic broadband users. This investment seeks increased users at the Wireless National User Facility to explore the capacity and quality of service effects from cyber or physical vulnerabilities and to develop innovative mitigation solutions for wireless communication systems. The research focuses on enhancing wireless security through experimental discovery and demonstration of how vulnerabilities and mitigation solutions can affect the capacity and quality of service of wireless systems that support critical infrastructure and emergency response. It also focuses on developing novel approaches for sharing spectrums among existing federal users and services while overcoming challenges in congestion management, managing spectrum sharing, developing new standards, and developing the testbed for broader applications.

- Delivering innovative products that enable defense, intelligence, and public safety: the objective of this investment is to develop proof-of-principle concepts demonstrating blast, ballistic, or energetic material innovations that protect the war fighter and critical infrastructure.
- Advancing the mission support center: the objective of this investment is to advance novel concepts for characterizing and demonstrating previously unidentified threats or threat indicators for critical infrastructure. Investments were also made for developing methodologies for translating threat information into metrics for prioritizing actions based on the significance of the type of threat, consequences if a threat is realized, or reduction of a threat by implementing protections.

### Call to Foster University Collaboration

Partnerships with universities, industry, and other national laboratories are encouraged. Two components of LDRD investments that encourage university collaboration are through investments in projects with the NUC and CAES universities as follows:

- NUC universities: investments in NUC universities focused on the following two areas: (a) building enhancements to Multi-Physics Object-Oriented Simulation Environment (MOOSE) and (b) development of nuclear hybrid energy systems. The focus of this investment is on a nuclear reactor heat source coupled to other clean energy generation systems (e.g., renewable sources) to produce electricity and to direct excess thermal and/or electrical energy to other industrial applications. This system could provide dispatchable power generation while accommodating a high penetration of intermittent renewable resources. Reactor designs that were considered included light water small modular reactor and advanced high-temperature reactor concepts. During FY 2016, INL invested in seven (\$1.4M) NUC related LDRD projects. All projects are part of the ANE portfolio.
- CAES universities: investments in CAES universities focused on addressing science and engineering challenges to help increase energy production through improvements in existing energy technologies and development of new ones, as well as an understanding of societal policy issues required for their implementation. Ideas were sought to address challenges in (a) nuclear energy and engineering; (b) materials science and engineering; (c) energy system design and analysis; (d) fossil carbon conversion; (e) geological systems and applications; (f) environment and sustainability; and (g) energy policy. During FY 2016, INL invested in two (\$600K) CAES-related LDRD projects, one project was part of the CED portfolio and another a part of the ANE portfolio.

### PROJECT AND INVESTMENT RELEVANCY TO PROGRAM OFFICES

In support of the objectives of the various DOE programs, FY 2016 projects had relevancy to the program offices shown in Table 2.

*Table 2. Primary relevancy and investments by program office.*

Program Office	Projects	Investment(\$K)
DOE-NE	54	\$13,009
Energy Efficiency and Renewable Energy	29	\$8,019
National Nuclear Security Administration	18	\$3,157
Fossil Energy	14	\$4,068
Electricity Delivery and Energy Reliability	25	\$5,439
Science	31	\$8,319

As reported in DOE's FY 2016 LDRD *Report to Congress*, indirects from DOE-NE programs contributed approximately \$7.53M to the LDRD Program at INL, which supported nuclear energy-related LDRD projects worth \$13.0M.

### REPORT ORGANIZATION

The remainder of this report is divided into three main sections, one for each of the three mission focus areas discussed above. These sections contain project write-ups that are arranged by project number. Each write-up contains (a) a description of the project, including its objectives and purpose; (b) a summary of the project's scientific or technical progress; (c) a statement of the benefits the project has provided to DOE's national security missions and, if applicable, the missions of other federal agencies; (d) a list of publications, presentations, invention disclosures, patents, and copyrights resulting from a project; (e) list of postdocs and interns supported through the project and who participated in delivering project outcomes; and (f) where applicable, list of university, industry, and national lab collaborators. Project summaries are categorized based on the three strategic areas. Appendices sections follow the individual project summaries. Appendix A lists agencies benefiting from INL's LDRD investments. Appendix B provides a summary list of all publications in FY 2015 and FY 2016, along with impact factors. Appendix C provides a summary list of university, industry, and national laboratory collaborations through LDRD projects and a summary of the scope of those collaborations. Appendix D provides a list of interns and postdocs and their university affiliations. Finally, Appendix E provides a list of patents for FY 2015 and FY 2016, as well as a brief description of the patents.





# Advancing Nuclear Energy

- 14-009—Development of an INL Capability for High-Temperature Flow, Heat Transfer, and Thermal Energy Storage with Applications in Advanced Small Modular Reactors, High-Temperature Heat Exchangers, Hybrid Energy Systems, and Dynamic Grid Energy Storage Concepts
- 14-010—Use of Linear Variable Differential Transformer-Based Methods to Detect Real-Time Geometry Changes during Irradiation Testing
- 14-025—Minor Actinide and Lanthanide Separations in Alternative Media
- 14-026—Multi-Scale Modeling on Delayed Hydride Cracking in Zirconium: Hydrogen Transport and Hydride Nucleation
- 14-031—Multi-Dimensional Multi-Physics Modeling of Fuel Behavior during Accident Conditions
- 14-037—Development of Advanced Nuclear Material Characterization Technology for Security Applications
- 14-041—Uranium Nitride – Uranium Silicide Composite Ceramic Fuel Production via Spark Plasma Sintering
- 14-045—End-to-End Radiation Detector Enhancements for Improved Safety and Security in Safeguarded Facilities
- 14-075—Development of Tools and Methodologies for Uncertainty Quantification and Validation for Multi-Physics Fuel Performance Simulation
- 14-098—Irradiation Effects of Uranium Dioxide
- 14-104—Development of a Multi-Physics Algorithm for Analyzing the Integrity of Nuclear Reactor Containment Vessels Subjected to Extreme Thermal and Overpressure-Loading Conditions
- 15-002—Experimental Scenarios of Adversity and Recovery in Aqueous Separations
- 15-013—Simulation Based Analysis of Procedures and Accident Management Guidelines
- 15-023—Development of Stochastic Three-Dimensional Soil Response Capability in MOOSE to Provide Design and Beyond Design Basis Seismic Motions for Nuclear Facilities
- 15-032—Development of New Method for High-Temperature Thermal Conductivity Measurements of Nuclear Materials
- 15-040—Acoustic Telemetry Infrastructure for In-Pile ATR and TREAT Monitoring
- 15-060—Development of Efficient TREAT Modeling with Graphite Data Improvement
- 15-094—Evaluation and Demonstration of the Integration of Safeguards, Safety, and Security by Design
- 15-141—Interfacing MOOSE Components to Enhance Capability
- 15-142—New In-Core Neutron Diagnostics
- 15-144—Investigation of Sonication-Assisted Electrolytic Reduction of Used Oxide Fuel in Molten Salt
- 15-145—Advanced Neutron and X-Ray Imaging at TREAT
- 16-003—Recycling of Tantalum-Containing Waste Materials to Recover Tantalum Metal
- 16-009—Change Detection System for Nuclear Applications

- 16-010—Development of a Fully Coupled Radiation Damage Production and Evolution Simulation Capability
- 16-013—Micromechanistic Approach and Critical Experiments for Quantitative Predictions of Delayed Hydride Cracking in Zirconium Alloys
- 16-017—Evaluation of Load-Following Capabilities of Existing and New Nuclear Power Reactors in the Grid with Large-Scale Renewable Energy Sources
- 16-026—Computationally Efficient Prediction of Containment Thermal Hydraulics Using Multi-Scale Simulation: Feasibility Study
- 16-033—Investigation of Gadolinium Nanocrystal Gels for Scintillator Use in Neutron Detection
- 16-036—Neutron Microscope to Enable High-Resolution Neutron Tomography at INL
- 16-040—Integration of Prognostic Techniques and Probabilistic Safety Assessment for Online Risk Monitoring
- 16-046—Development of a Synergistic Approach to Study Irradiated Materials Using Coupled Experiments and Simulation
- 16-050—Stress Corrosion Cracking Testing in Supercritical Carbon Dioxide
- 16-055—Capability Extension for Multi-Scale, Multi-Application Development within the Multi-Physics Object-Oriented Simulation Environment
- 16-058—Predicting Radiation-Induced Microstructural Change via Implementation and Validation of Multi-Scale Cluster Dynamics in MOOSE
- 16-070—Characterization of Neutron Beamlines at NRAD
- 16-071—Evaluation of Advanced Digital Neutron Imaging Systems for PIE of Nuclear Fuel
- 16-085—Production of Fluoroanion Targets for Accelerator Mass Spectrometry
- 16-096—Supporting Operator Performance and Situation Awareness in Highly Automated Nuclear Power Plants
- 16-098—Nuclear Nonproliferation Applications of  $^{14}\text{C}$  Analyses by Accelerator Mass Spectrometry
- 16-129—Application of Radioactive Isotope Dilution Technique to Measurement for Molten Salt Mass for the Electrochemical Recycling Process
- 16-149—In-Core Qualification of Developmental Instrumentation
- 16-187—Micro-Scale Technique to Evaluate Grain Boundary Cohesion of Irradiated Alloys
- 16P6-003FP—Phenomena Identification and Ranking Table Technique Applied to the MEGA-POWER Heat Pipe Reactor Concept

## **14-009—Development of an INL Capability for High-Temperature Flow, Heat Transfer, and Thermal Energy Storage with Applications in Advanced Small Modular Reactors, High-Temperature Heat Exchangers, Hybrid Energy Systems, and Dynamic Grid Energy Storage Concepts**

James E. O'Brien,<sup>1</sup> Piyush Sabharwall,<sup>1</sup> and Su Jong Yoon<sup>1</sup>

### **General Project Description**

This project was focused on design, development, and analysis of effective and robust high-temperature heat transport systems and working fluids to support successful deployment of advanced high-temperature reactor systems for both power generation and nonelectric applications. These heat transport systems typically make use of an intermediate heat transport loop (IHTL) designed to isolate the process heat application from the primary reactor coolant. Advanced high-temperature, high-pressure heat exchangers are needed at each end of the IHTL. Printed circuit heat exchangers (PCHEs) are strong candidates for this application due to their very high power density, requiring much less material per unit of heat duty compared to conventional shell-and-tube heat exchangers and their high-pressure capability. Candidate high-temperature working fluids for the IHTL include molten fluoride salts. Molten salts have excellent high-temperature heat transfer and chemical stability characteristics.

### **Summary**

The initial phase of this project (FY 2014) was focused on the development of a conceptual design for a high-temperature, multi-fluid, multi-loop test facility to support heat transfer, flow, materials, and thermal energy storage research for nuclear and nuclear-hybrid applications. In its initial configuration, this facility will include a high-temperature helium (He) loop, a liquid salt IHTL, and a hot water/steam loop. The three loops will be thermally coupled through an intermediate heat exchanger and a secondary heat exchanger. Research topics to be addressed include the characterization and performance evaluation of candidate compact heat exchangers such as PCHEs at prototypical operating conditions, flow and heat transfer issues related to core thermal hydraulics in advanced He-cooled and salt-cooled reactors, and evaluation of corrosion behavior of new cladding materials and accident-tolerant fuels for light water reactors at prototypical conditions. Based on its relevance to advanced reactor systems, the new capability has been named the Advanced Reactor Technology Integral System Test (ARTIST) facility. The facility will be located at INL.

The second and third years of this project (FY 2015 and FY 2016) were focused on supporting technology development and analysis. Detailed computational fluid dynamic (CFD) evaluation of the heat transfer and pressure-drop performance of PCHEs was completed (see Figure 1), including parametric studies to determine the effects of the various geometric flow configurations available with PCHEs. Parameters considered in the analysis include channel hydraulic diameter, header design, channel shape, channel streamwise configuration (zigzag or straight), working fluid, and flow rate. For the zigzag configurations, additional parameters were considered, including zigzag angle and type of corner (rounded or abrupt). The CFD work was performed in collaboration with The Ohio State University. The Ohio State University has provided experimental data derived from the operation of PCHEs in the university's high-temperature helium loop, including two straight-channel PCHEs and one zigzag-channel PCHE. Performance data obtained from testing of these heat exchangers were compared to CFD predictions for code validation. As part of the validation process, we have developed a new methodology for analysis of experimental heat exchanger data, accounting for extraneous heat losses. The parametric studies completed under this task are new and unique and have led to several publications.

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<sup>1</sup> Idaho National Laboratory

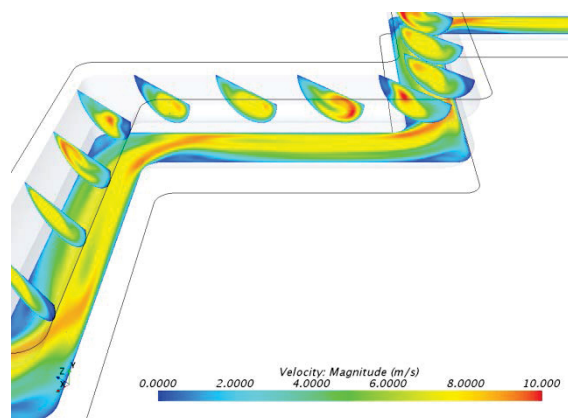


Figure 1. Velocity magnitude distributions in PCHE zigzag channel flow.

A new experimental laboratory capability for small-scale preparation and purification of fluoride salt mixtures has also been developed. This capability is needed to support the final design and operation of the ARTIST facility. The prepared salt mixtures will be tested for thermodynamic properties such as melting point, heat capacity, and thermal conductivity. The salts will also be used in a pot experiment in which liquid salts will be transferred from one pot to another to validate high-temperature trace heating and flow measurement methods. Purification of fluoride salt mixtures is accomplished by hydrofluorination, which removes the moisture/oxide impurities and other contaminants such as chlorine and sulfur. Hydrofluorination is accomplished by HF/H<sub>2</sub>/He sparging of the molten salt mixture at ~450°C. A glovebox has been outfitted to support these salt-related activities. Additionally, the laboratory gas-monitoring system has been upgraded to detect HF, and a ventilated gas safety cabinet has been installed to safely house the HF gas. New salt-preparation and transfer vessels have been fabricated from Alloy 201(Ni). Several samples of FLiNaK have been prepared and purified and are ready for additional analysis, as shown in Figure 2.



Figure 2. FLiNaK ingot and glassy carbon crucible.

## Benefits to DOE

This project has laid the foundation for development of a new thermal-hydraulic experimental capability at INL to support the DOE-NE Advanced Reactor Technology program and nuclear-hybrid energy systems research. We are moving forward immediately with the ARTIST facility with FY 2017 funding from the DOE-NE nuclear-hybrid energy systems program. This loop will be deployed at the INL Energy Systems Laboratory and will be collocated with other hybrid energy system components, including thermal energy storage and hydrogen production capabilities and dynamic response instrumentation. The final detailed design, procurement, and assembly of the high-pressure, high-temperature water loop portion high-pressure water loop will operate at



pressurized water reactor conditions, with research objectives related to fuel cladding corrosion, accident-tolerant fuels, natural circulation studies, and hybrid energy systems.

There is an increased level of interest within DOE and industry in the use of molten salts for high-temperature reactor coolants and heat transport fluids. The small-scale molten salt preparation and purification facility developed under this project will support development of the molten salt flow loop portion of the ARTIST facility, as well as basic thermophysical property measurements and corrosion studies. Programmatic support for INL molten salt work is expected in FY 2017. In addition, an FY 2017 Nuclear Energy University Program proposal (with the University of Michigan) has been submitted to support molten salt corrosion research.

## Publications

### Conference Papers

- Chen, M., J. E. O'Brien, S. Yoon, R. N. Christensen, P. Sabharwall, and X. Sun, 2016, "Thermal-Hydraulic Performance of a High-Temperature Zigzag-Channel Printed Circuit Heat Exchanger," *8th International Topical Meeting on High Temperature Reactor Technology*, Las Vegas, Nevada, November 6 through 10, 2016.
- O'Brien, J. E., P. Sabharwall, and S. Yoon, 2014, "Development of a Multi-Loop Flow and Heat Transfer Facility for Advanced Nuclear Reactor Thermal Hydraulic and Hybrid Energy System Studies," *2014 ASME International Congress and Exposition*, Montreal, November 14 through 20, 2014, Paper IMECE2014-39057.
- O'Brien, J. E., P. Sabharwall, and S. Yoon, 2014, "A Multi-Purpose Thermal Hydraulic Test Facility for Support of Advanced Reactor Technologies," *ANS 2014 Winter Meeting and Nuclear Technology Expo*, Anaheim, California, November 9 through 13, 2014, Paper 11705.
- Sabharwall, P., J. E. O'Brien, and S. Yoon, 2015, "Experimental Facility for Development of High-Temperature Reactor Technology: Instrumentation Needs and Challenges," *International Congress on Advances in Nuclear Power Plants (ICAPP2015)*, Nice, France, May 3 through 6, 2015, Paper 15066.
- Yoon, S., J. E. O'Brien, and M. Chen, 2016, "CFD-Aided Development of Nusselt Number and Friction Factor Correlations for Laminar Flow in Semi-Circular Zigzag Channels of Printed Circuit Heat Exchangers," in preparation, September 2016.
- Yoon, S., K. Wegman, J. E. O'Brien, P. Sabharwall, and X. Sun, 2016, "Thermal-Hydraulic Performance of Printed Circuit Heat Exchangers: CFD Analysis with Experimental Validation," *8th International Topical Meeting on High Temperature Reactor Technology*, Las Vegas, Nevada, November 6 through 10, 2016.
- Yoon, S., K. Wegman, J. E. O'Brien, P. Sabharwall, and Xiaodong Sun, 2016, "An Analytical Methodology to Evaluate Thermal-Hydraulic Performance of Compact Heat Exchangers, Accounting for Heat Loss," *2016 International Congress on Advances in Nuclear Power Plants (ICAPP2016)*, San Francisco, California, April 17 through 20, 2016, Paper 15814.

### Journal Articles

- Sabharwall, P., J. E. O'Brien, S. Yoon, and X. Sun, 2015, "Experimental Facility for Development of High-Temperature Reactor Technology: Instrumentation Needs and Challenges," *European Physical Journal of Nuclear Science and Technology*, Vol. 1, No. 14, December 2015.

### External Report

- O'Brien, J. E., P. Sabharwall, and S. Yoon, 2014, Strategic Need for Multi-Purpose Thermal Hydraulic Loop for Support of Advanced Reactor Technologies, INL/EXT-14-33300, September 2014.



## **Presentations**

- O'Brien, J. E., 2014, "Nuclear Energy in the Context of Today's Energy Challenges," *Nuclear Engineering Seminar Series*, The Ohio State University and University of Cincinnati (video link), Columbus, Ohio, March 26, 2014.
- O'Brien, J. E., 2015, "Development of a Multi-Loop Flow and Heat Transfer Facility for Advanced Nuclear Reactor Thermal Hydraulic and Hybrid Energy System Studies," *INL Annual Program Review*, DOE Germantown Office, September 22, 2015.
- O'Brien, J. E., 2015 "Development of a Multi-Loop Flow and Heat Transfer Facility for Advanced Nuclear Reactor Thermal Hydraulic and Hybrid Energy System Studies," presentation to Terrestrial Energy, INL, July 15, 2015.
- O'Brien, J. E., 2015, "Development of a Multi-Loop Flow and Heat Transfer Facility for Advanced Nuclear Reactor Thermal Hydraulic and Hybrid Energy System Studies," presentation to Transatomic Power, INL, June 1, 2015.
- O'Brien, J. E., 2015, "Development of a Multi-Loop Flow and Heat Transfer Facility for Advanced Nuclear Reactor Thermal Hydraulic and Hybrid Energy System Studies," *Nuclear Science and Technology Peer Review*, INL, May 20, 2015.
- O'Brien, J. E., 2015, "Development of a Multi-Loop Flow and Heat Transfer Facility for Advanced Nuclear Reactor Thermal Hydraulic and Hybrid Energy System Studies," *LDRD Annual Review*, INL, May 12, 2015.
- Sabharwall, P. and S.-J. Yoon, 2016, "Advanced Reactor Technology Integral System Test (ARTIST) Facility," presentation to Oak Ridge National Laboratory researchers, September 21, 2016.

## **Interns and Postdocs**

Interns: Kevin Wegman, Minghui Chen, Xiaoqin Zhang, and In Hun Kim

Postdoc: Su-Jong Yoon

## **Collaborations**

University: Ohio State University

Industry: Technology Insights

## 14-010—Use of Linear Variable Differential Transformer-Based Methods to Detect Real-Time Geometry Changes during Irradiation Testing

Kurt Davis,<sup>1</sup> Richard Skifton,<sup>1</sup> John Crepeau,<sup>2</sup> and Steinar Solstad<sup>3</sup>

### General Project Description

New materials are always being considered for fuel, cladding, and structures in advanced and existing nuclear reactors. These materials can undergo various dimensional changes during irradiation. Currently, in the United States, these changes are measured by the “cook and look” method (i.e., repeatedly irradiating a specimen for a specified period of time and then removing it from the reactor for evaluation). The time and labor required for removing, examining, and returning irradiated samples for each measurement makes this approach very expensive. In addition, the techniques provide limited data, as well as handling may disturb the phenomena of interest. Therefore, in-pile detection of changes in geometry is sorely needed to understand real-time behavior during irradiation testing of fuels and materials in high flux U.S. material and test reactors (MTRs). In that regard, a linear variable differential transformer (LVDT) was modified and utilized to measure the diameter of a simulated fuel rod with calibrated, known dimensions. Under the design parameter that pressure in the reactor’s test loop drives the sample/fuel rod, an innovative hydraulic piston was designed to keep a seal on the loop fluid medium while still permitting axial motion of the apparatus.

### Summary

Over the life of this project (i.e., Fiscal Year 2014 through Fiscal Year 2016), the LVDT-based methods for measuring diameter of a simulated fuel rod have been deemed successful. These proof-of-concept measurements will have broad applicability to many U.S. MTRs. The measurements relied on withdrawing or inserting the simulated fuel rod at a moderately slow constant velocity (i.e., about 2.2 mm/s). Using *constant* velocity, this instrument integrates with time to attain the position of the measurement. This greatly reduces the number of feedthroughs needed in the vessel head, reducing the current LVDT sensor down to only three feedthroughs.

The proof-of-concept was achieved during year two of the project, and it showed repeatability of the measurements within  $\pm 3\text{ }\mu\text{m}$  of the calibrated diameter and within  $\pm 90\text{ }\mu\text{m}$  of the location of the diameter measurement.

During the third year, the focus turned to having a leak tight, metal-on-metal hydraulic ram. This allowed for the reactor loop pressure to drive the sample media outward and be hydraulically reinserted, while avoiding melting and/or activating the seal. This capability was shown tremendous support during the final LDRD poster session of the project.

Future work involves testing the LVDT in an autoclave environment. This will lead into specimen testing within the ATR test loop.

### Benefits to DOE

This project provides benchmark studies on the way to real-time monitoring of dimensional changes during irradiation testing in MTRs. This capability does not currently exist at U.S. MTRs. This project also supports opportunities for DOE Office of Naval Reactors (e.g., ATR), DOE-NE (e.g., Nuclear Sciences User Facility, Next Generation Nuclear Plant, Fuel Cycle Research and Development, Nuclear Energy Enabling Technology, and

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<sup>1</sup> Idaho National Laboratory

<sup>2</sup> University of Idaho

<sup>3</sup> Halden Reactor Project, Institute for Energy Technology, Norway

Light Water Reactor Sustainability), and industry (e.g., Electric Power Research Institute, Westinghouse, and General Electric).

## **Publications**

Davis, K. L., D. L. Knudson, J. L. Rempe, J. C. Crepeau, and S. Solstad, 2015, "Design and Laboratory Evaluation of Future Elongation and Diameter Measurements at the Advanced Test Reactor," *Nuclear Technology*, Vol. 191, MS# NT 14-60R.

Davis, K., D. L. Knudson, J. C. Crepeau, and S. Solstad, 2015, "Measurement of Diameter Changes During Irradiation Testing," *9th International Topical Meeting on Nuclear Plant Instrumentation, Control, and Human Machine Interface Technologies (NPIC&HMIT)*, Paper #12432.

Rempe, J., D. Knudson, J. Daw, T. Unruh, B. Chase, K. Davis, R. Schley, J. Palmer, C. White, and K. Condie, 2014, *Status Report on Efforts to Enhance Instrumentation to Support Advanced Test Reactor Irradiations*, INL/EXT-13-30427, Idaho National Laboratory.

Rempe, J. L., D. L. Knudson, J. E. Daw, T. C. Unruh, B. M. Chase, K. L. Davis, A. J. Palmer, and R. S. Schley, 2014, "Advanced In-Pile Instrumentation for Materials Testing Reactors," *IEEE Transactions on Nuclear Science*, Vol. 61, No. 4, pp. 1984-1994.

## **Presentations**

Davis, K., D. L. Knudson, J. C. Crepeau, and S. Solstad, 2015, "Measurement of Diameter Changes During Irradiation Testing," *9th International Topical Meeting on Nuclear Plant Instrumentation, Control, and Human Machine Interface Technologies (NPIC&HMIT)*, San Diego, California.

## **Invention Disclosures, Patents, Copyrights**

It is anticipated that two invention disclosure requests will be submitted from the work performed in this project.

## **Interns and Postdocs**

Postdoc: Richard Skifton

## **Collaborations**

University: University of Idaho

National Laboratories: Institute for Energy Technology/Halden Reactor Project

## 14-025—Minor Actinide and Lanthanide Separations in Alternative Media

Mitchell Greenhalgh<sup>1</sup> and R. Scott Herbst<sup>1</sup>

### General Project Description

Since its inception, the nuclear industry, specifically the reprocessing of irradiated nuclear fuels, has realized that separation of minor actinides (MA) (e.g., americium and curium) from lanthanides (LN) and their subsequent recycle into new reactor fuel would greatly reduce waste generated by reprocessing. However, separation of MA from LN represents one of the most daunting challenges faced by modern day separation scientists and is rivaled in complexity only by isotopic separations. LN/MA separation is difficult due to the remarkably similar chemical behavior between the LN series (which includes the elements of lanthanum through lutetium and yttrium) and MA. The ultimate goal of this project is to develop and test efficient solvent extraction flowsheets and equipment for separation of MA from LN elements in alternative media (i.e., chloride, sulfate, phosphate, or thiocyanate). Traditional nuclear processing separations have focused on nitrate media (e.g.,  $\text{HNO}_3$ ), and the process equipment (notably constructed from stainless steel) is compatible only with the nitrate media. Industrially, separation and purification of the LN species typically uses chloride media (e.g.,  $\text{HCl}$ ) and different extractants and diluents than those indigenous to the nuclear industry. Ironically, most of these industrially proven systems have not been evaluated for MA/LN separation despite the inordinately similar chemistry of these elements in solution. Laboratory experiments will be performed to establish the fundamental separations behavior of MA and LN in alternative systems. Experimentally acquired data will be used to develop and test process flowsheets for the most promising separation schemes. Additionally, chemically compatible solvent extraction equipment that is suitable for use in a glove box or hot cell will be designed, procured, and tested for the new flowsheets with actual radionuclides of interest in the laboratory hoods at tracer levels.

### Summary

Laboratory experiments for determining the fundamental separation characteristics and behaviors of MA and LN using alternative extractants and non-traditional aqueous media were performed throughout the life of this project. The extractants that were investigated include PC88A, Cyanex 572, Cyanex 923, tetraoctyldiglycolamide, and octyl(phenyl) diisobutylcarbamoyl methylphosphine oxide. The aqueous systems examined include chloride, nitrate, sulfate, phosphate, and thiocyanate. A metal recycle in the scrub section, similar to techniques commonly employed in the rare earth industry, was investigated as well. Although a separation factor of 50 was obtained for europium/americium with the PC88A/chloride system, separation factors of the light lanthanides (i.e., cerium) from americium were quite low. The results from these laboratory tests did not identify any improvements to the separation factors between americium and cerium under the conditions evaluated.

In addition to separations testing, 32 stages of small, laboratory-scale mixer-settler equipment constructed of a Kynar® polymer were obtained from Metalextraktion AB of Sweden. This equipment is chemically compatible with all proposed alternative media in the project and is also the most radiation-resistant plastic available. They have a maximum throughput of 166 mL/min and a total solution hold up of only 600 mL per stage. The equipment can be operated in a variety of process configurations, which were to be defined by the concurrent separations evaluations. The entire 32-stage system only requires approximately 12 linear ft of bench space, making it ideal for testing in a space-limited radiological environment. Initial hydraulic evaluations of the mixer-settler equipment were performed to define the operating parameters that would achieve the desired separations (i.e., maximum flowrates, mixing speeds, and mass transfer efficiencies). Additional extraction, scrubbing, and stripping tests were performed in multiple stages of the mixer-settlers using the PC88A/chloride system. These tests were performed to evaluate performance of the equipment for the various sections of a

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<sup>1</sup> Idaho National Laboratory

flowsheet and to gain operational experience with the equipment. The results of this testing demonstrated that equipment will perform as desired for any flowsheet configuration that would be identified. Although an acceptable separation of MA from the light LNs was not achieved, this equipment provides INL with a unique separations capability that exists nowhere else in the United States.

## Benefits to DOE

The chemistry and engineering expertise developed and separations processes examined are directly relevant to INL's nuclear fuel cycle mission, especially development of separation capabilities that can be applied to processes different from the norm. The equipment and engineering developments related to process flowsheets are also directly relevant to future nuclear fuel cycle separations. Furthermore, the unique separations capabilities and equipment obtained in this project represent the only location in the United States where such work can be performed. This unique capability has the potential to secure new funding from a wide range of DOE and Department of Defense offices with interests in nuclear fuel cycle or other similar separations (e.g., rare earths).

## Publications

### ***Presentations***

Lyon, K. L., M. R. Greenhalgh, and R. S. Herbst, 2015, "Minor Actinide and Lanthanide Separations in Alternative Media," *39<sup>th</sup> Annual Actinide Separations Conference*, Salt Lake City, Utah, May 18 through 21, 2015.

Lyon, K. L., M. R. Greenhalgh, R. D. Tillotson, and M. M. Warner, 2016, "Application of Industrial Perspectives to the Nuclear Fuel Cycle," *40<sup>th</sup> Annual Actinide Separations Conference*, San Diego, California, May 23 through 26, 2016.

## Interns and Postdocs

Intern: Justin McAlister

## 14-026—Multi-Scale Modeling on Delayed Hydride Cracking in Zirconium: Hydrogen Transport and Hydride Nucleation

Yongfeng Zhang,<sup>1</sup> Xianming Bai,<sup>1</sup> and Simon Phillpot<sup>1</sup>

### General Project Description

Formation of hydrides in zirconium (Zr)-based cladding leads to embrittlement and is therefore a critical material-science issue affecting fuel pin integrity in light water reactors (Allen et al. 2012). While hydrides have been widely characterized experimentally, the transport of hydrogen (H) and the mechanisms for hydride nucleation are not fully understood. The objectives of this project are to use atomic-scale modeling tools to (1) predict H diffusivity, (2) assess the thermodynamic driving forces for hydride precipitation, and (3) explore the mechanisms for hydride nucleation. The modeling tools, results, and data collected from atomic-scale research will be useful for upper-scale modeling of hydride formation and delayed hydride cracking during used fuel disposition.

### Summary

This LDRD is in its third and final year. Significant progress has been made in exploring the diffusion of H, thermodynamics of hydride phases, and nucleation mechanism of  $\gamma$ -hydride. The research outcome has led to six presentations at leading conferences, including two invited talks at conferences, one delivered at the Materials, Science, and Technology 2016 meeting and the other to be delivered at the Materials Research Society 2017 spring meeting. The results have been featured in a journal article, with a few more articles expected. The project has supported a Ph.D. student from the University of Florida. The modeling tools, results, and data collected are being used by several directly funded programs. Although this LDRD is ending, funding has been captured from the DOE-NE Advanced Modeling and Simulation program to continue research in the same area. In summary, this LDRD is successful in many strategic project metrics, including impacting science and engineering, creating program growth opportunities, and building strategic collaborations.

The diffusion of H in  $\alpha$ -Zr governs the kinetics of H uptake and delayed hydride cracking. To obtain a systematic understanding and reliable diffusivity data, an accelerated kinetic Monte Carlo method was developed and parameterized by density functional theory calculations. The accelerated kinetic Monte Carlo method is efficient in calculating diffusivity, with excellent agreement with experimental results, as shown in Figure 1. Also, the Ishioka model (Ishioka and Koiwa 1985) has been demonstrated to be accurate for H diffusivity in  $\alpha$ -Zr, and a correction using the Oriani model (Oriani 1970) is sufficient for that in Zircaloy, provided that all hopping rates are available. The kinetic Monte Carlo method can be directly applied to other hexagonal close packed (hcp) metals and will be extended for cases involving stress fields and lattice defects. The results have been summarized in a journal manuscript under review. Positive comments have been received and the revised version will be submitted soon.

Various hydride phases have been identified by modeling and experiments, with a systematic thermodynamic assessment yet to be accomplished. Assisted by the use of the density functional theory, an analysis was conducted of the driving forces for hydride precipitation. As shown in Figure 2, at 0 K, only hcp Zr and tetragonal ZrH<sub>2</sub> are thermodynamically stable. The  $\gamma$ -ZrH lies slightly above the phase separation line and is thus on the borderline of stable and metastable. For the contribution of configurational entropy, at finite temperature,  $\delta$ -hydride becomes stable, sharing similar stability with hypo-stoichiometric  $\epsilon$ -hydride. This explains the frequent observation of  $\delta$ -hydride in experiments. The lattice parameters and elastic constants of various hydrides are also calculated (Table 1) to provide necessary data for thermomechanical analysis of hydride precipitates in a cladding matrix.

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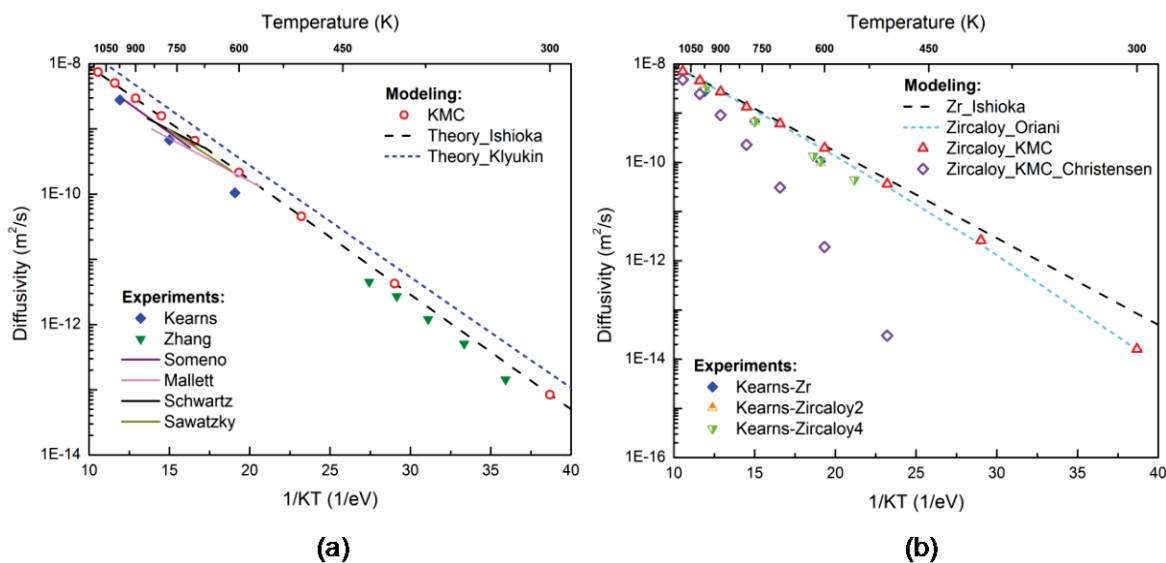


Figure 1. H diffusivity in (a)  $\alpha$ -Zr and (b) Zircaloy.

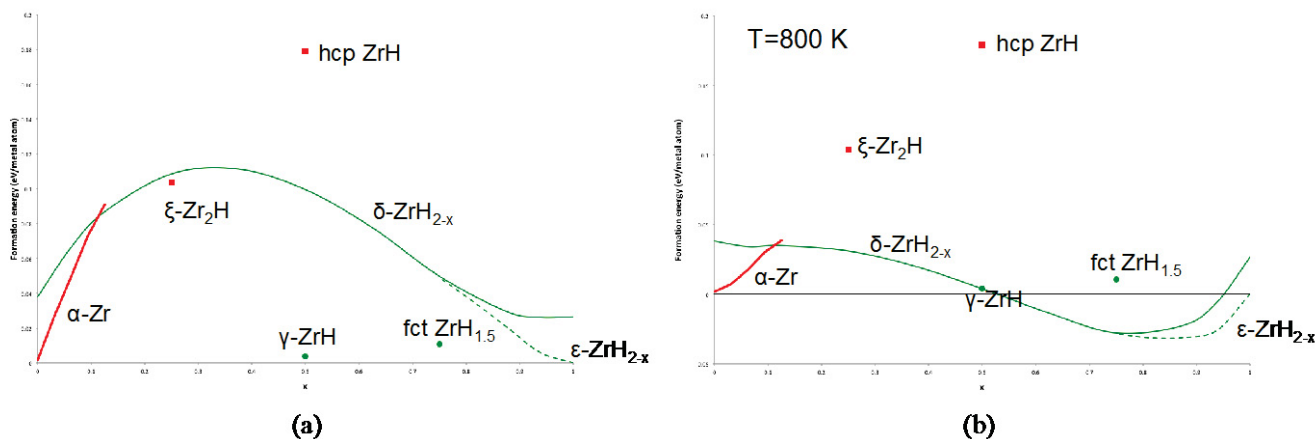


Figure 2. Formation enthalpies of hydrides at (a) 0 K and (b) 800 K as functions of H content. Lower enthalpy means higher stability. H solid solution is represented by  $\alpha$ -Zr (red).

In Zircaloy claddings, the most relevant hydride is the  $\delta$ -hydride, which previous experiment results suggest forms from  $\gamma$ -hydride. Using the charge-optimized-many-body potential, a nucleation path for homogeneous  $\gamma$ -hydride formation in  $\alpha$ -Zr, from solid solution to coherent  $\zeta$ -hydride and then  $\gamma$ -hydride, has been revealed by molecular dynamics simulations, as shown in Figure 3. At finite temperatures, the  $\zeta$ -hydride goes through an hcp to face-centered-cubic (fcc) transformation, forming a mixture of  $\gamma$ -hydride and  $\alpha$ -Zr, which can further grow into a  $\gamma$ -hydride cluster. It was further found that the hcp to fcc transformation is thermodynamically favored with a negligible barrier. The mechanism observed here is consistent with literature results and is believed to be relevant at reactor operating temperatures. Having provided insightful results, the charge-optimized-many-body potential is being further optimized to better describe all hydride phases.

Table 1. Calculated equilibrium lattice parameters and elastic constants of Zr hydrides. For comparison, results from previous experimental and theoretical studies are also shown in parentheses.

Compound	Structure	<i>a</i>	<i>b</i>	<i>c</i>	<i>c</i> <sub>11</sub>	<i>c</i> <sub>22</sub>	<i>c</i> <sub>33</sub>	<i>c</i> <sub>12</sub>	<i>c</i> <sub>23</sub>	<i>c</i> <sub>13</sub>	<i>c</i> <sub>44</sub>	<i>c</i> <sub>55</sub>	<i>c</i> <sub>66</sub>	<i>B</i>	<i>G</i>
ZrH <sub>0.5</sub>	Cubic	4.66 (4.66 <sup>a</sup> )			120			96			58			104	31
ZrH	Tetragonal	4.59 (4.58 <sup>a</sup> ) (4.59 <sup>b</sup> ) (4.58 <sup>d</sup> ) (4.60 <sup>e</sup> )		5.03 (5.04 <sup>a</sup> ) (5.02 <sup>b</sup> ) (5.02 <sup>d</sup> ) (4.97 <sup>e</sup> )	123 (122 <sup>d</sup> )		183 (183 <sup>d</sup> )	116 (116 <sup>d</sup> )	95 (98 <sup>d</sup> )		49 (48 <sup>d</sup> )		61 (61 <sup>d</sup> )	115 (113 <sup>b</sup> ) (116 <sup>d</sup> )	27 (25 <sup>b</sup> ) (25 <sup>d</sup> )
ZrH	Orthorhombic	4.19	4.94	5.05	99	160	192	112	97	85	25	63	57	104	30
ZrH <sub>1.5</sub>	Tetragonal	5.04 (5.04 <sup>a</sup> )		4.26 (4.25 <sup>a</sup> )	166		119	132	103		57		38	120	33
ZrH <sub>2</sub>	Tetragonal	5.01 (5.01 <sup>a</sup> ) (5.00 <sup>b</sup> ) (5.03 <sup>c</sup> ) (5.01 <sup>d</sup> ) (4.98 <sup>e</sup> )		4.43 (4.44 <sup>a</sup> ) (4.46 <sup>b</sup> ) (4.41 <sup>c</sup> ) (4.42 <sup>d</sup> ) (4.45 <sup>e</sup> )	172 (166 <sup>c</sup> ) (166 <sup>d</sup> )		154 (146 <sup>c</sup> ) (149 <sup>d</sup> )	146 (141 <sup>c</sup> ) (149 <sup>d</sup> )	106 (107 <sup>c</sup> ) (109 <sup>d</sup> )		31 (31 <sup>c</sup> ) (27 <sup>d</sup> )		68 (61 <sup>c</sup> ) (56 <sup>d</sup> )	133 (130 <sup>b</sup> ) (130 <sup>c</sup> ) (133 <sup>d</sup> )	31 (24 <sup>b</sup> ) (29 <sup>c</sup> ) (25 <sup>d</sup> )

<sup>a</sup>From Domain et al. 2002.

<sup>b</sup>From Christensen et al. 2015.

<sup>c</sup>From Togo and Tanaka. 2015.

<sup>d</sup>From Zhang et al. 2011.

<sup>e</sup>From Zuzek et al. 1990.

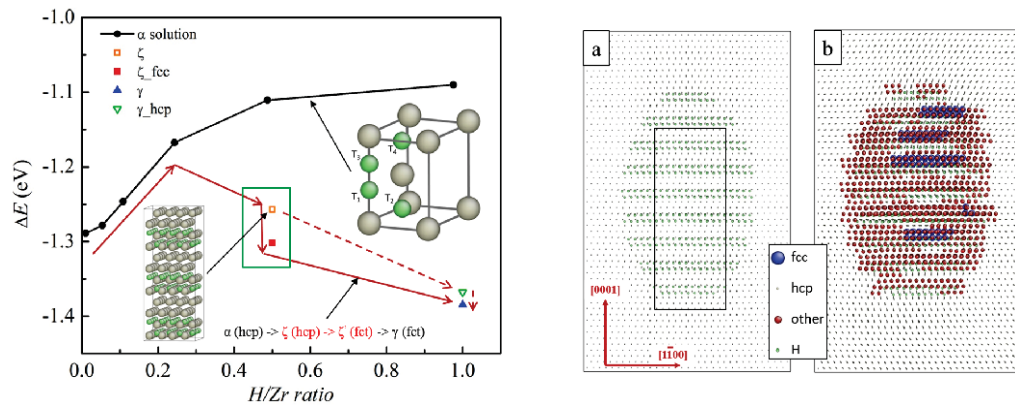


Figure 3. Left: H solution energy as a function of H/Zr ratio. Right: atomic configurations showing the structural transformation of an embedded  $\zeta$ -phase cluster to a  $\gamma$ -hydride nucleus at 600 K.

## Benefits to DOE

The research outcome of this project largely advanced our understanding of hydride formation, including H transport and hydride nucleation. The success of this project helps to evaluate the detrimental effects of hydrides as they relate to decreased periods of used fuel storage and thus the risks they pose to nuclear safety and energy security. The modeling tools developed and the data generated can be used in other LDRD projects and directly funded programs. The research outcome has led to multiple conference presentations and journal publications. It

benefits DOE's mission by impacting science and engineering, building strategic collaborations, and establishing thought leadership by providing scientific understanding, collaborating with a university professor, and supporting a Ph.D. student.

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## Publications

- Jiang, C. and Y. Zhang, "Ab initio study on the phase stability of Zr hydrides," to be submitted.
- Zhang Y., C. Jiang, and X. Bai, "Anisotropic hydrogen diffusion in  $\alpha$ -Zr and Zircaloy predicted by accelerated kinetic Monte Carlo simulations," *Nature Scientific Report*, accepted.
- Zhang, Y., X. Bai, J. Yu, M. R. Tonks, M. Noordhoek, and S. Phillpot, 2016, "Homogeneous hydride nucleation pathway in Zr: molecular dynamics simulations with the charge-optimized-many-body potential," *Acta Materialia*, Vol. 111, pp. 357–365.

## Presentations

- Bai, X., Y. Zhang, and M. R. Tonks, 2016, "Computer Modeling of Hydrogen and Oxygen Transport during Zirconium Corrosion," *TMS 2016 Meeting*, Nashville, Tennessee, February 14 through 18, 2016.
- Zhang, Y., W. Xu, P. Millett, and Y. Zhu, 2014, "Inter-facet Vacancy Diffusion and Void Nucleation in hcp Metals," *Materials Research Society 2014 Fall Meeting*, Boston, Massachusetts, November 30 through December 5, 2014.
- Zhang, Y., X. Bai, M. Noordhoek, and S. Phillpot, 2015, "Molecular Dynamics Simulations of Zr/Zr-hydride Interface with the Charge Optimized Many Body (COMB) Potential," *TMS 2015 Meeting*, Orlando, Florida, March 15 through 19, 2015.
- Zhang, Y., C. Jiang, X. Bai, J. Yu, M. Fullarton, M. Noordhoek, and S. Phillpot, 2016, "Understanding Hydride Formation in Alpha-Zr at the Atomic Scale," *MS&T 2016 Meeting*, Salt Lake City, Utah, October 23 through 27, 2016.

Zhang, Y., X. Bai, J. Yu, M. R. Tonks, M. Noordhoek, and S. Phillpot, 2016, “Molecular dynamics simulations on homogeneous hydride nucleation in alpha-Zr,” *TMS 2016 Meeting*, Nashville, Tennessee, February 14 through 18, 2016.

Zhang, Y., C. Jiang, X. Bai, S. B. Biner, and S. Phillpot, 2017, “Atomistic to mesoscale understanding of hydride formation in alpha-Zr,” *Materials Research Society Spring 2017 Meeting*, Phoenix, Arizona, April 17 through 21, 2017.

## Interns and Postdocs

Postdoc: Michele Fullarton

## 14-031—Multi-Dimensional Multi-Physics Modeling of Fuel Behavior during Accident Conditions

Richard Williamson,<sup>1</sup> Jason Hales,<sup>1</sup> Giovanni Pastore,<sup>1</sup> Stephen Novascone,<sup>1</sup> and Wen Jian<sup>1</sup>

### General Project Description

Developing computational tools for reliably predicting thermo-mechanical behavior and the lifetime of nuclear fuel rods during abnormal reactor events is essential from both safety and economic standpoints. Fuel performance codes able to cope with accident analysis are needed for safety assessments and design purposes, as well as for development of accident-tolerant fuel concepts. The present research aims to develop an advanced tool for improved analysis of accident fuel behavior relative to traditional modeling, leveraging the unique capabilities of INL's fuel performance code BISON. The focus is on simulation of loss-of-coolant accidents (LOCAs), which is very relevant following the Fukushima accident.

### Summary

Several limitations exist in current fuel performance codes, including simplistic 1.5-dimensional approximation, loosely coupled physics, and need of separate codes for normal operation and accident analysis. The current project focuses on achieving significant advances relative to state-of-the-art accident fuel analysis, building on INL's multi-dimensional, multi-physics, fully coupled fuel performance code BISON. The outcome is a tool that is able to model integral fuel rod behavior during both normal reactor operation and accident conditions, beyond the limitations of traditional analysis and with improved accuracy.

During the project, BISON was expanded with capabilities to reproduce all main phenomena involved in light water reactor fuel rod behavior during LOCA conditions. In particular, models are now available in BISON for (1) transient fission gas release coupled with gaseous swelling, (2) high-temperature steam-cladding oxidation, (3) Zircaloy phase transformation, (4) hydrogen behavior in Zircaloy, (5) Zircaloy high-temperature creep, and (6) cladding burst failure. Furthermore, to simulate cladding oxidation with moving material layer (i.e., metal and oxide) boundaries, a novel moving finite element formulation has been developed using the extended finite element method.

Significant validation of the expanded BISON code was performed. Some results from a first-time, three-dimensional simulation of a cladding ballooning-burst test under LOCA conditions are illustrated in Figure 1. A visual inspection image of a burst-failed fuel rod is also included. The results demonstrate capability to reproduce experimentally observed behavior with the new tool (i.e., expanded BISON) developed during the project. This includes three-dimensional effects such as azimuthal temperature variations, which play an important role in fuel rod behavior during LOCAs (e.g., localized burst) and cannot be captured with traditional one point five and two-dimensional codes. Validation of the extended code through quantitative comparison of results against an extensive experimental dataset was also performed. For example, predictions of burst failure for 31 separate effects cladding tests are compared to experimental data in Figure 2 (left). Moreover, integral LOCA experiments from the Halden Reactor Project have been analyzed with BISON to validate capability to accurately model integral fuel rod behavior during LOCAs. Figure 2 (right) shows results comparing calculated and experimental rod pressure evolution and time to burst during the LOCA transient. Results demonstrate satisfactory accuracy of BISON predictions.

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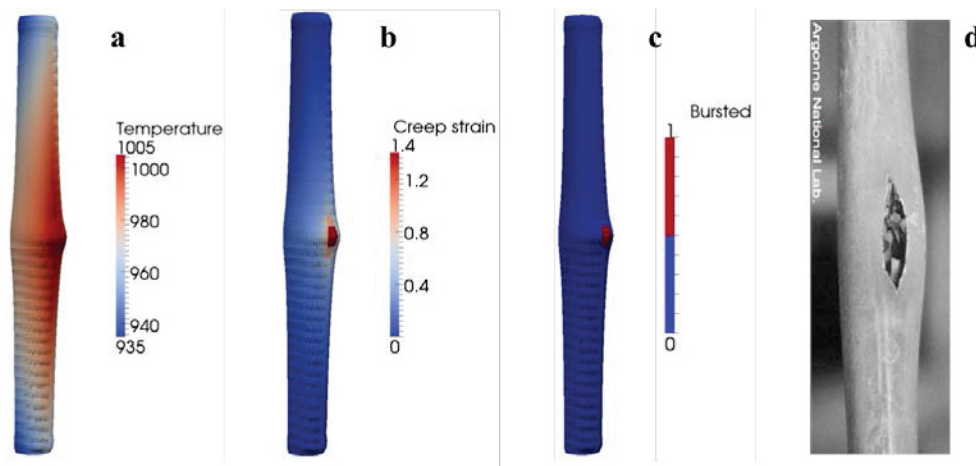


Figure 1. Contour plots for a BISON three-dimensional simulation of a cladding LOCA test, including temperature (a), creep strain (b), and burst failure area (c). Visual inspection of a burst-failed fuel rod following LOCA is also shown (d).

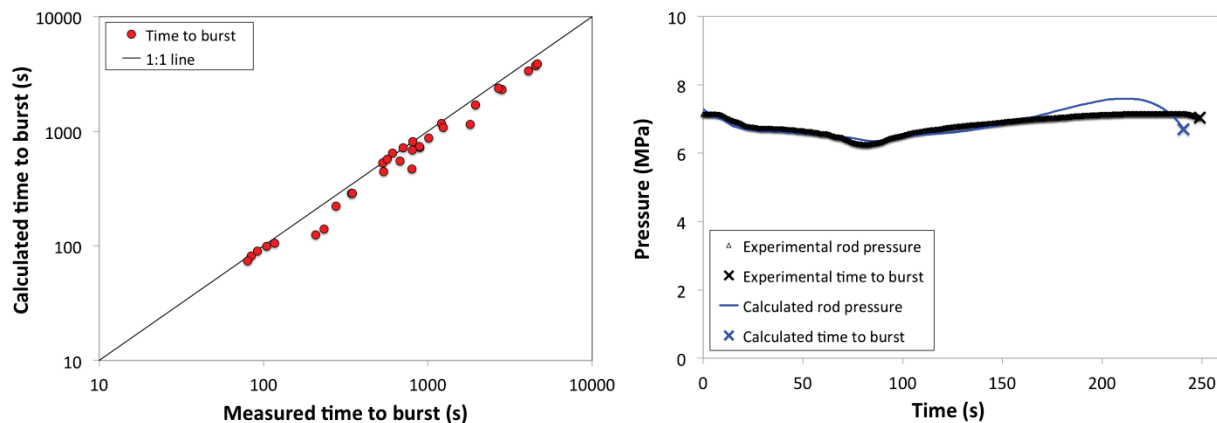


Figure 2. Validation of BISON for fuel rod burst failure during LOCAs. Calculations are compared to experimental data for 31 separate-effects cladding burst tests (left) and the Halden integral fuel rod experiment IFA-650.10 (right).

This work has been presented at several international meetings. Also, this project led to DOE participation in the International Atomic Energy Agency-sponsored Coordinated Research Project on Fuel Modeling under Accident Conditions. Moreover, important international collaborations relevant to this project have been established with the Institute for Transuranium Elements (European Commission, Germany), Politecnico di Milano University (POLIMI, Italy), and the Organization for Economic Cooperation and Development's Halden Reactor Project (Norway). These collaborations involved several student internships at INL and an assignment at Halden Reactor Project for an INL BISON developer on analysis of Halden LOCA tests.

## Benefits to DOE

This current project directly benefits the DOE-NE mission by addressing one of today's most critical nuclear energy concerns—design basis accidents. Following the Fukushima accident, and given the experimental evidence from programs such as the Halden Reactor Project, understanding LOCAs and reducing their likelihood



are key concerns. This work advances the state-of-the-art of fuel modeling by providing the ability to explore accident behavior in a novel and improved way. This project represents a strong addition to current nuclear fuel modeling efforts at INL and further demonstrates INL's developing excellence in modeling and simulation science.

## Publications

- Barani, T., E. Bruschi, D. Pizzocri, G. Pastore, P. Van Uffelen, R. L. Williamson, and L. Luzzi, 2016, "Analysis of transient fission gas behaviour in oxide fuel using BISON and TRANSURANUS," *Journal of Nuclear Materials*, in press.
- Chakraborty, P., M. R. Tonks, and G. Pastore, 2014, "Modeling the influence of bubble pressure on grain boundary separation and fission gas release," *Journal of Nuclear Materials*, Vol. 452, pp. 95-101.
- Pastore, G., D. Pizzocri, J. D. Hales, S. R. Novascone, D. M. Perez, B. W. Spencer, R. L. Williamson, P. Van Uffelen (ITU), and L. Luzzi (POLIMI), 2014, "Modelling of Transient Fission Gas Behaviour in Oxide Fuel and Application to the BISON Code," *Proceedings of the Enlarged Halden Programme Group Meeting*, Røros, Norway, September 7 through 12, 2014.
- Pastore, G., S. R. Novascone, R. L. Williamson, J. D. Hales, B. W. Spencer, and D. S. Stafford, 2015, "Modeling of Fuel Behavior during Loss-of-Coolant Accidents using the BISON Code," *Proceedings of the Reactor Fuel Performance Meeting – Top Fuel 2015*, Zurich, Switzerland, September 13 through 17, 2015.
- Pastore, G., R. L. Williamson, S. R. Novascone, B. W. Spencer, and J. D. Hales, 2016, "Modelling of LOCA Tests with the BISON Fuel Performance Code," *Proceedings of the Enlarged Halden Programme Group Meeting*, Fornebu, Norway, May 8 through 13, 2016.
- Pizzocri, D., C. Rabiti, L. Luzzi, T. Barani, P. Van Uffelen, and G. Pastore, 2016, "PolyPole-1: An accurate numerical algorithm for intra-granular fission gas release," *Journal of Nuclear Materials*, Vol. 478, pp. 333-342.
- Stafford, D. S., 2015, "Multidimensional simulations of hydrides during fuel rod lifecycle," *Journal of Nuclear Materials*, Vol. 466, pp. 362-372.

## Presentations

- Pastore, G., D. Pizzocri, J. D. Hales, S. R. Novascone, D. M. Perez, B. W. Spencer, R. L. Williamson, P. Van Uffelen, and L. Luzzi, 2014, "Modelling of Transient Fission Gas Behaviour in Oxide Fuel and Application to the BISON Code," *Enlarged Halden Programme Group Meeting*, Røros, Norway, September 7 through 12, 2014.
- Pastore, G., S. R. Novascone, R. L. Williamson, J. D. Hales, B. W. Spencer, and D. S. Stafford, 2015, "Modeling of Fuel Behavior during Loss-of-Coolant Accidents using the BISON Code," *Reactor Fuel Performance Meeting – Top Fuel 2015*, Zurich, Switzerland, September 13 through 17, 2015.
- Pastore, G., R. L. Williamson, and J. D. Hales, 2016, "Status report on INL contribution to FUMAC with the BISON code," *Second FUMAC Research Coordination Meeting*, Vienna, Austria, May 30 through June 2, 2016.
- Pastore, G., R. L. Williamson, S. R. Novascone, B. W. Spencer, and J. D. Hales, 2016, "Modelling of LOCA Tests with the BISON Fuel Performance Code," *Enlarged Halden Programme Group Meeting*, Fornebu, Norway, May 8 through 13, 2016.

Williamson, R. L., G. Pastore, S. R. Novascone, J. D. Hales, B. W. Spencer, and D. M. Perez, 2014, “Modeling LOCA behavior with the BISON fuel performance code,” *20<sup>th</sup> QUENCH Workshop*, Karlsruhe, Germany, November 11 through 13, 2014.

### **Invention Disclosures, Patents, Copyrights**

CW-16-11, Tracking No. 4175 (underway), “PolyPole program for the calculation of intra-granular fission gas release,” Software Disclosure.

### **Interns and Postdocs**

Postdoc: Wen Jiang

## 14-037—Development of Advanced Nuclear Material Characterization Technology for Security Applications

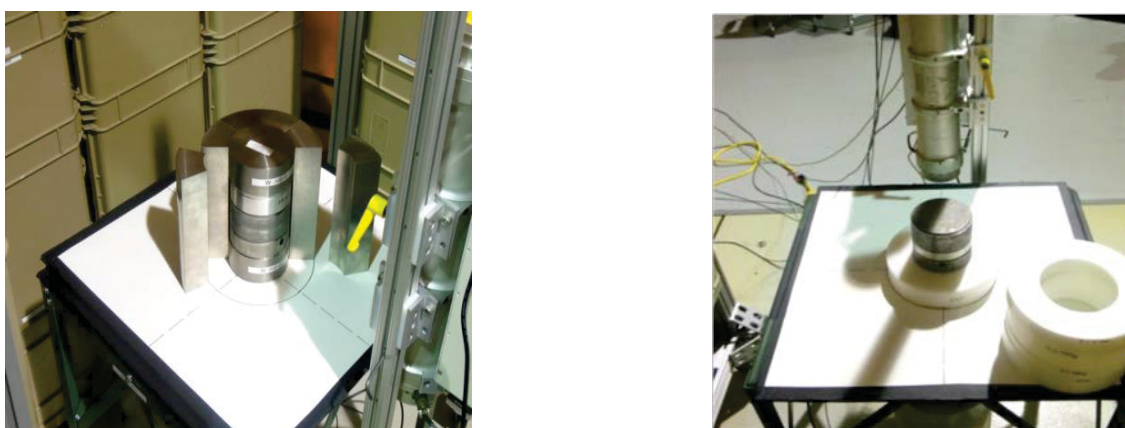
David L. Chichester,<sup>1</sup> James T. Johnson,<sup>1</sup> Mathew T. Kinlaw,<sup>1</sup> Scott J. Thompson,<sup>1</sup> and Scott M. Watson<sup>1</sup>

### General Project Description

Emerging nuclear nonproliferation, counterproliferation, and forensic challenges facing the United States require new technological developments in the area of radiation measurement. Often, research programs addressing these areas focus on one type of sensor and one particular observable. It is our hypothesis that the integrated use of orthogonal radiation measurement concepts for detecting and analyzing observables will improve our ability to characterize these materials versus separate uses of these technologies. This project is exploring new sensor combinations that can be used in support of national objectives for detecting nuclear and radiological materials.

### Summary

Traditional active neutron interrogation (ANI) signatures (such as prompt neutron die-away analysis, delayed neutron analysis, and delayed gamma-ray analysis) have each been shown useful as diagnostic methods for detecting shielded special nuclear material (SNM) or for verifying the absence of SNM within inspected items. However, comparatively less work has taken place to explore the use of these measurement techniques for characterizing assemblies of shielded SNM to determine attributes such as SNM mass, the composition of SNM, or the configuration of SNM. In contrast, time-correlated neutron analysis methods (such as thermal-neutron coincidence counting and thermal-neutron multiplicity counting) are proven measurement techniques for characterizing SNM.



*Figure 1. Test setup for interrogating highly enriched uranium (HEU) assemblies. The photograph on the left shows two HEU disks in contact with each other within a 5-cm thick tungsten shell (some parts removed to see the inside). The photograph on the right shows the same two HEU disks, this time with a 2-cm thick aluminum disk in between them; 5-cm thick polyethylene rings are placed to the side prior to being lowered around the HEU.*

Experiments were performed at INL's Zero Power Physics Reactor facility to explore the potential for integrating traditional ANI signatures from SNM with time-correlated neutron analysis methods. This work used an array of neutron detector modules specially designed at INL for use in ANI, along with a newly-acquired list mode data acquisition system. This new data acquisition system is capable of individually recording the relative time of

<sup>1</sup> Idaho National Laboratory

arrival of neutron detection events in the array in relation to an arbitrary reference time (for passive measurements and for interrogation using steady-state neutron sources) or in relation to synchronization time stamps from a pulsed electronic neutron generator. Measurements were performed using varying configurations of INL's MARVEL test assembly of metallic, HEU. The maximum HEU mass used in these trials was 14.35 kg. The material was used bare, reflected with 5 cm of polyethylene, and reflected with 5 cm of tungsten (W). Neutron multiplication levels ranged from 1.8 (5.49 kg of HEU, bare) to 8.8 (14.35 kg of HEU, reflected with 5 cm of polyethylene). Photographs of two test configurations are shown in Figure 1.

Example results from these experiments are shown in Figure 2. These three examples show cases where (a) the HEU mass is the same, (b) the HEU multiplication level (determined empirically and through simulation) is the same, and (c) both the HEU mass and multiplication level are the same. For each case, considering both the die-away and the  $\beta$ -delayed neutron signals, clear differentiation between the different scenarios is possible within a few minutes. In contrast, for these three cases, it is unlikely that either passive gamma-ray spectrometry or passive gross or time-correlated neutron counting alone could be relied on to independently detect the scenario differences.

In addition to this work area, this project has also carried conceptual investigations to examine the use of hand-carried radiation detection instrumentation for the purpose of surveying the post-blast environment of a radiological dispersal device, with the goal of developing instrumentation and methodology to reconstruct the pre-blast radiological dispersal device source term. This work included developing hand-held detector systems and a telemetry toolkit capable of relaying real-time dose field data to a remotely located command post. Additionally, new tools were developed to visualize these data.

## Benefits to DOE

This project provides benefit to DOE by advancing the science and engineering principles for development of advanced diagnostic instruments and methods capable of characterizing shielded assemblies of SNM, in particular HEU. New characterization capabilities being developed in this project have relevance across a number of DOE focus areas, including criticality safety, used-fuel storage, nuclear facility operations, nuclear nonproliferation, safeguards, arms control, and treaty verification. Work executed in this project in relation to post-blast radiological dispersal device site characterization is relevant to DOE emergency response programmatic activities.

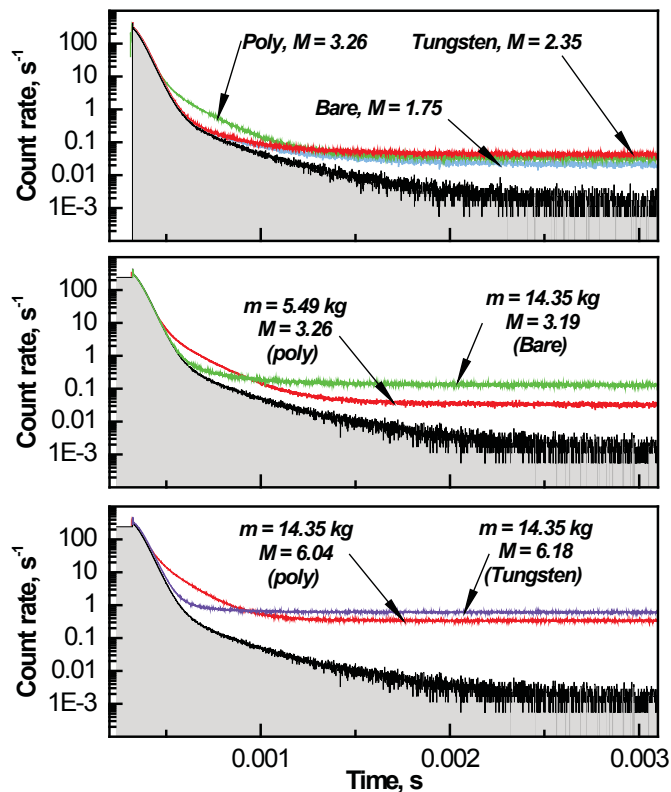


Figure 2. Three figures comparing the inter-pulse neutron signatures observed from differing assemblies of HEU. Top (same mass): three distinct signals for cases where the HEU mass is constant and the reflector multiplication values vary. Middle (same multiplication): two cases where the multiplication is nearly the same, but the mass and reflector vary. Bottom (same mass and multiplication): two cases where mass and multiplication are nearly identical, but the reflector varies.

## Publications

Chichester, D. L., S. J. Thompson, M. T. Kinlaw, J. T. Johnson, J. L. Dolan, M. Flaska, and S. Pozzi, 2015, "Statistical Estimation of the Performance of a Fast Neutron Multiplicity System for Nuclear Material Accountancy," *Nuclear Instruments and Methods in Physics Research A*, Vol. 784, pp. 448-454.

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Chichester, D. L., M. T. Kinlaw, S. M. Watson, J. Kalter, E. Miller, and W. Noonan, 2014, "Experiments and Simulations of the Use of Time-Correlated Thermal Neutron Counting to Determine the Multiplication of an Assembly of Highly Enriched Uranium," Institute of Electrical and Electronics Engineers Nuclear Science Symposium, Seattle, Washington, November 2014.

Chichester, D. L., S. J. Thompson, M. T. Kinlaw, J. T., Johnson, J. Dolan, M. Flaska, and S. Pozzi, 2014, "Statistical Estimation of the Performance of a Fast-Neutron Multiplicity System for Nuclear Material Accountancy," 2014 Symposium on Radiation Measurements and Applications (SORMA XV), Ann Arbor, Michigan, June through 12, 2014.

Chichester, D. L., S. J. Thompson, M. T. Kinlaw, J. T. Johnson, M. J. Dolan, M. Flaska, and S. Pozzi, 2014, "Statistical Estimation of the Performance of a Fast Neutron Multiplicity System for Nuclear Material Accountancy," 2014 Symposium on Radiation Measurements and Applications (SORMA XV), Ann Arbor, Michigan, June 9 through 12 , 2014.

## Interns and Postdocs

Interns: Jay D. Hix, Thomas V. Holschuh, and Charles S. Sosa

## Collaborations

Industry: PHDs Company

## 14-041—Uranium Nitride – Uranium Silicide Composite Ceramic Fuel Production via Spark Plasma Sintering

Robert C. O'Brien,<sup>1</sup> Jason Harp,<sup>1</sup> and Paul A. Lessing<sup>1</sup>

### General Project Description

The goal for the work performed during FY 2016 was to continue the feasibility assessment of fabricating uranium mononitride (UN) – uranium disilicide ( $\text{U}_3\text{Si}_2$ ) composites using the spark plasma sintering (SPS) process and extend the initial research and development to explore net shape production of prototypical fuel pellets. The specifically desired microstructure was to create microencapsulation of UN particles by amorphous  $\text{U}_3\text{Si}_2$ . A volume fraction of 60% UN in a 40%  $\text{U}_3\text{Si}_2$  matrix was examined to provide a sufficient uranium atom density in the composite such that these compacts may be used as a replacement for  $\text{UO}_2$  fuel pellets in existing and future power reactors. An additional composition of 30% UN in a 70%  $\text{U}_3\text{Si}_2$  matrix was also fabricated in FY 2016 to provide greater compatibility as a like-for-like drop-in replacement for  $\text{UO}_2$  with respect to reactivity, but with greater protection of the UN components.

### Summary

The SPS process uses rapid pulses of direct current at voltages of up to 10 V and typical current densities on the order of 103 to 104 A/cm<sup>2</sup> to sinter powdered materials held within a conductive die. The sintering mechanisms that take place during SPS are highly dependent on the electrical properties of the materials being processed. For electrically conductive materials, the process promotes initial rapid joule heating at high resistance points across the microstructure. For non-conductive materials, the rapid joule heating of the surrounding die results in rapid heating of the powdered materials enclosed via thermal conduction, which, in turn, promotes grain boundary diffusion and surface diffusion.

One of the fundamental objectives for FY 2016 was to further demonstrate the feasibility of net shape production of composite fuel pellets to prototypical geometries. In order to accomplish this, initial work in FY 2015 was performed to design, fabricate, and develop a new configuration of SPS tooling that would produce final pellet geometries with the highest possible density when completely closed. Improvements to the design of this tooling were made in FY 2016 to better facilitate assembly. This tooling has successfully demonstrated repeatable production of prototypical pellets without any requirement for refurbishment or repair. Figure 1 shows a photograph of a prototypical pellet produced by this tooling. An invention disclosure for this tooling is being drafted and submitted for patent application review.

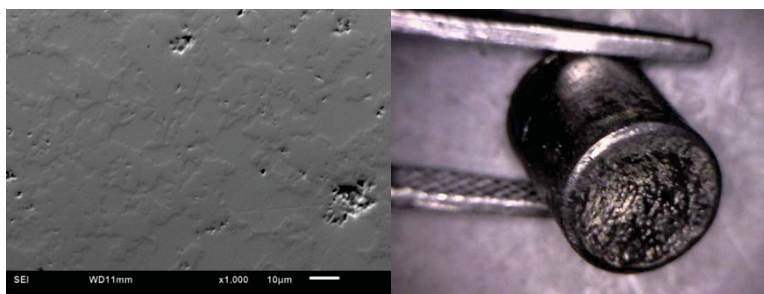


Figure 1. (left) Scanning electron microscope image of a prototypical fuel pellet (right) that was sectioned for microscopy. Pullout due to polishing is seen in the localized regions of the image.

<sup>1</sup> Idaho National Laboratory



Preliminary results have successfully demonstrated the feasibility of producing sintered composites of UN -  $U_3Si_2$  to high density via the SPS process. The work performed in FY 2014 identified that a peak sintering temperature of 1300°C was required to produce an amorphous  $U_3Si_2$  phase around a dispersion of UN particles. The work in FY 2014 also identified minor challenges with the formation/decomposition of  $U_3Si_2$  to form localized elemental silicon phases during high-temperature sintering. In FY 2015, this challenge was solved through application of a controlled cooling schedule. Evidence conclusively shows that the UN- $U_3Si_2$  composite was densified by liquid phase sintering where the  $U_3Si_2$  phase surrounds every grain of UN. This is illustrated in Figure 1. The more corrosion-resistant phase (i.e.,  $U_3Si_2$ ) has melted and completely surrounded each of the UN grains. Liquid-phase sintering enhancement requires a molten second phase that wets and spreads on powder surfaces within a compact. Figure 1 (left and right) show clear evidence that all four stages of liquid-phase sintering (i.e., wetting, rearrangement, dissolution-re-precipitation, and agglomeration) have occurred and that elemental silicon phases can be significantly reduced by means of controlled cooling. In FY 2016, pellet fabrication with aged powder feedstock was demonstrated. This is essential for industrial scale-up activities for accident-tolerant fuels because vendors are likely to store powder feedstock between pellet batches. The results showed that SPS can successfully fabricate net-shaped pellets with powders that are over 2 years old.

X-ray diffraction analysis was performed in FY 2016 on composite pellets fabricated by SPS. The results of the bulk x-ray diffraction analyses reveal high-purity UN phases surrounded by  $U_3Si_2$ . No minor alternative phases of the uranium-silicon system were observed, indicating the capability for SPS to produce uniform matrices with the bulk stoichiometry provided in the feedstock powder.

During FY 2016, autoclave corrosion experiments have been run using cross sections of prototypical pellets at the Materials and Fuels Complex's Fuels and Applied Science Building. The experiment emulates the conditions of reflood of the reactor vessel with pressurized water reactor coolant (with prototypical chemistry) over pellets that have been exposed from failed fuel claddings. The corrosion rate and pellet mass trend as a function of exposure time to boiling portable water are measured throughout these experiments and will be published in a journal article upon conclusion of these tests in FY 2016.

## Benefits to DOE

Fuel pellets composed of a composite matrix of  $U_3Si_2$  and UN are of interest as an accident-tolerant, high-density, uranium-dispersion fuel form for light water reactors. Specifically, these pellets might be a direct substitute for metallic fuels or  $UO_2$  fuels in the existing light water reactor fleet. Also, the improved thermal conductivity of the  $U_3Si_2$ -UN fuel increases the safety margin of the reactor. This project has demonstrated that prototype pellets can be fabricated by the SPS net-shape process. This is likely to support future DOE program needs in producing unique test specimens of varying compositions for specific applications without the need for complicated re-tooling between fabrication campaigns.

## Publications

O'Brien, R. C., P. A. Lessing, and J. Harp, 2016, "Fabrication of UN- $U_3Si_2$  Composite Fuel Pellets via Spark Plasma Sintering," *Journal of Nuclear Materials*, article in draft.

## Presentations

O'Brien, R. C. and P.A. Lessing, 2015, "Fabrication of UN- $U_3Si_2$  Composites via Spark Plasma Sintering," *LDRD Review Meeting*, March 13, 2015.

O'Brien, R. C. et al., 2016, "Fabrication of Accident Tolerant UN- $U_3Si_2$  Composite Fuel Pellets by Spark Plasma Sintering," *INL Annual LDRD Poster Session*, CAES Building, August 30, 2016.

## **Invention Disclosures, Patents, Copyright**

INL IDR in draft September 2016

## 14-045—End-to-End Radiation Detector Enhancements for Improved Safety and Security in Safeguarded Facilities

S. J. Thompson,<sup>1</sup> C. S. Sosa,<sup>1</sup> D. L. Chichester,<sup>1</sup> D. Shy,<sup>1</sup> and M. M. Grinder<sup>1</sup>

### General Description of the Project

The project objective is demonstrating that radiation detection systems (such as portal monitors) located at safeguarded facilities can be significantly enhanced through an end-to-end improvement of the scintillation-based detection equipment used within these systems. The use of advanced simulation and modeling techniques in unison with benchmark laboratory testing will allow us to optimize the physical layout and light collection properties associated with the detectors' scintillating crystals for heightened sensitivity to radioactive material. System security can also be enhanced through application of INL-developed data protection methods to transmitted data, reducing the risk of monitor spoofing, data corruption, or eavesdropping. These improvements will not only strengthen efforts to safeguard nuclear materials within facilities but also have the potential to enhance radiation safety measures that protect a site's workforce.

### Summary

The third and final year of this project focused on exploring the effects of geometric shape and coating materials on organic scintillator performance. Several detector crystal shapes were obtained for testing and to provide benchmark values for the Geant4-based simulation parameter study, which spans a complete parameter space of crystal shape and design. Examples of these detectors are shown in Figure 1. The scintillating volumes were coated with absorbing (black), diffuse (white), and specular (mirror-like) coverings, providing a second dimension to the parameter space. An example of how these coatings affect the light output from a 2-in. cylinder of pulse-shape-discriminating plastic irradiated with 662 keV gamma is also provided in Figure 1.

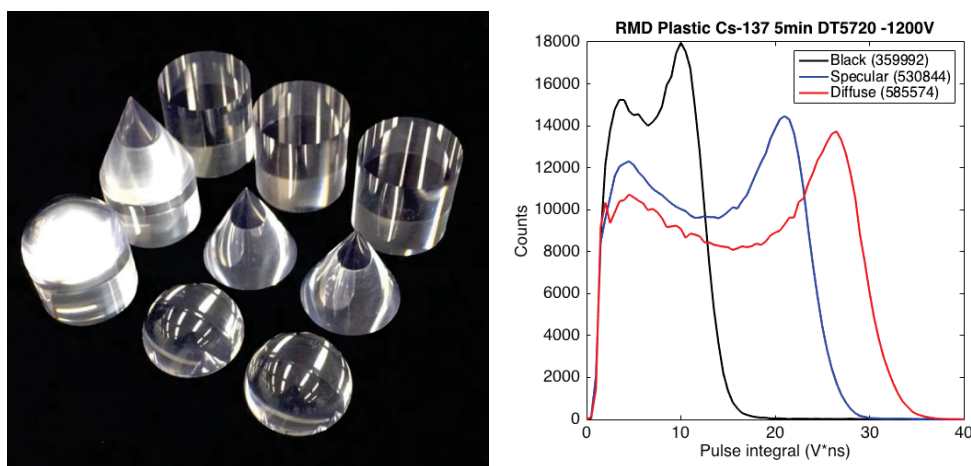


Figure 1. (left) A photograph of the EJ-200 organic scintillator shapes studied during this project. (right) An example of the effect of scintillator coating on spectral response.

Energy response was measured with a Compton backscatter-gated technique that uses a secondary high-efficiency detector positioned at 180 degrees from the organic scintillator to monitor for gamma that hits the organic scintillator and bounces back. This technique filters out the Compton continuum of the measured spectrum to

<sup>1</sup> Idaho National Laboratory

provide a single light peak at the energy of a 180-degree Compton backscatter. A before-and-after example of this filtering process is shown in Figure 2. This work will continue at the University of Michigan as a Ph.D. thesis.

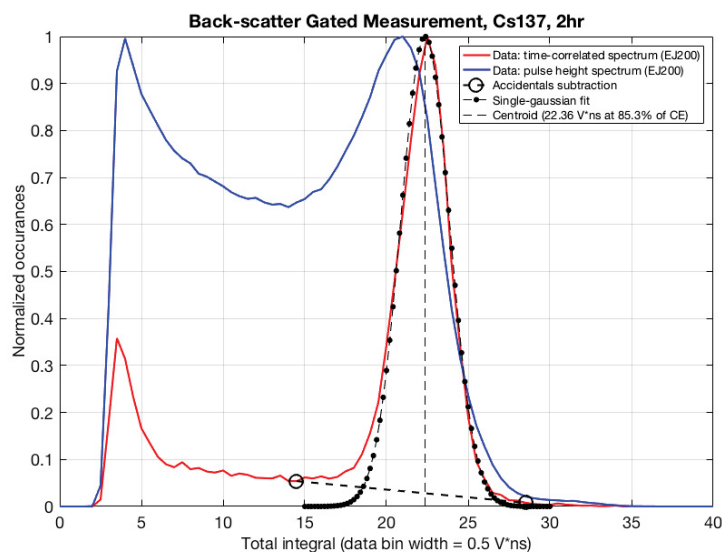


Figure 2. An example of the Compton backscatter filtering technique used to obtain an energy resolution value from the organic scintillators.

## Benefits to DOE

Less expensive, high-fidelity sensors will allow for increased confidence in radiation detection for nuclear nonproliferation and counter-proliferation measurements related to advanced safeguards, nuclear forensics, emergency response, and nuclear material detection.

## Publications

### Presentations

Sosa, C. S., 2016, "Optimization of Organic Scintillators for Nuclear Nonproliferation Applications," *Nuclear Nonproliferation Seminar Series*, Idaho National Laboratory, September 22, 2016.

## Interns and Postdocs

Interns: Mara Grinder, Charles Sosa, and Daniel Shy

## Collaborations

University: University of Michigan

## 14-075—Development of Tools and Methodologies for Uncertainty Quantification and Validation for Multi-Physics Fuel Performance Simulation

Congjian Wang,<sup>1</sup> Cristian Rabiti,<sup>1</sup> Richard L. Williamson,<sup>1</sup> Anil Prinja,<sup>2</sup> Paul W. Talbot,<sup>2</sup> Andrea Alfonsi,<sup>1</sup> Mandelli Diego,<sup>1</sup> and Joshua J. Cogliati<sup>1</sup>

### General Project Description

This project is focused on delivering a mathematical and software framework to perform uncertainty propagation for multi-physics simulations involving both neutronics and fuel performance simulations. Fuel performance analysis is characterized by long computational times that are often further complicated by nonlinear phenomena. Additionally, many of the parameters used in nuclear fuel behavior models are characterized by large uncertainties. Multi-physics simulations also introduce uncertainties from other models not used in typical fuel modeling. In particular, coupling neutronics calculations with fuel performance calculations exacerbates computational expense by injecting nonlinearity and increasing the dimensionality of the input space. Given the complexities of sophisticated modeling, the estimation of uncertainties characterizing fuel performance simulation is a difficult challenge. The goal of this project is to meet this challenge.

### Summary

During the first year of this project, the classical uncertainty quantification methodologies were implemented for fuel performance simulations (i.e., BISON). In more detail, an interface between RAVEN and BISON was created to obtain the capability of performing common statistical analyses (such as mean, sigma, skewness, kurtosis, and correlation coefficients). In addition, a prototypical BISON case was used to characterize the dimensionality and nonlinearity of a fuel performance analysis.

During the last 2 years of this project, the coupling between RAVEN and multi-physics simulations (i.e., MAMMOTH) have been extended. The advanced uncertainty propagation methodologies, including stochastic collocation with generalized polynomial chaos expansions and high dimensional model reduction, have been implemented. Advanced adaptive algorithms have been developed for both stochastic collocation with generalized polynomial chaos expansions and high dimensional model reduction. While these collocation-based techniques represent state-of-the-art methodologies for uncertainty quantification, they still are not capable of practically addressing the high dimensionality of the input space (e.g., cross sections) when reactor physics (i.e., neutron transport and nuclide depletion) are also accounted for in fuel performance analysis. To overcome these challenges, we have developed and implemented the multi-step input reduction technique using both input-input correlations and input-output correlations. This technique seeks to remove the linear combination of input parameters that have no impact on the figures of merit for the responses of interest. When the effective dimensionality is low, the collocation-based reduced order models (ROMs) that are fast for evaluating and acting as surrogate to the original expensive models are constructed. These ROMs can then be used to perform sensitivity and uncertainty analysis with very low computational cost. To verify the accuracy and efficiency of these methodologies, many analytic models and neutronics and fuel performance models have been tested. In addition, we have implemented many supporting functionalities in the RAVEN framework such as multi-variate distributions, code interfaces, post-processors, and ROMs. Figure 1 compares the accuracy and efficiency between the traditional Monte Carlo method and the advanced methods integrated with the multi-step input reduction technique.

<sup>1</sup> Idaho National Laboratory

<sup>2</sup> University of New Mexico

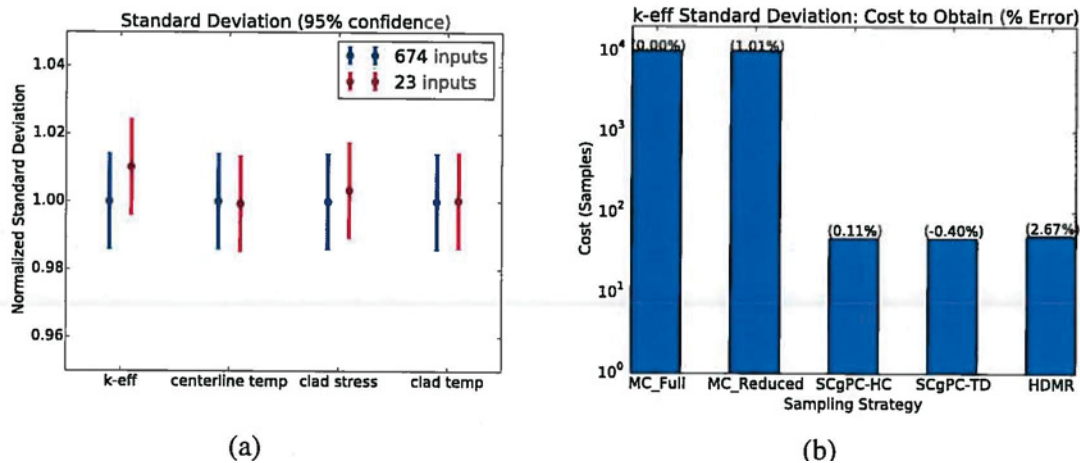


Figure 1. Uncertainty analysis using multi-step input reduction (a) combined with the advanced uncertainty propagation methodologies (b) for coupled neutronics and fuel performance simulations.

Methodologies developed during research and development have been presented at several international conferences and have opened the way for advanced uncertainty quantification on a scale that was previously inaccessible. Techniques developed in this work are being used in RAVEN both at INL and at multiple universities and national laboratories. The project has provided an opportunity for doctoral research in cooperation with the University of New Mexico and currently supports a post-doctoral appointment at INL.

## Benefits to DOE

Benefits from the introduction of high-fidelity simulation tools have been experienced in many other fields, and, during the last decade, DOE has invested in creation of such tools for nuclear energy applications. The nuclear sector is highly regulated; therefore, as a consequence, it exhibits a strong inertia in the adoption of new technologies. It is clear that without efficient quantification of a new tool's uncertainties, it will not be licensed in the nuclear industry. This is precisely the point addressed by this project and success in this project will allow the industrial deployment of several million dollars of already-invested money, generating a true benefit to the nuclear industry.

## Publications

- Rabiti, C., J. Cogliati, G. Pastore, and A. Alfonsi, 2015, "Fuel Reliability Analysis Using BISON and RAVEN," *International Topical Meeting on Probabilistic Safety Assessment and Analysis*, Sun Valley, Idaho, April 26 through 30, 2015.
- Talbot, P. W. and A. Prinja, 2014, "Sparse Grid Stochastic Collocation Uncertainty Quantification Convergence for Multigroup Diffusion," *Transactions of American Nuclear Society*.
- Talbot, P. W., A. K. Prinja, and C. Rabiti, 2015, "High Density Model Reduction Uncertainty Quantification for Multigroup Diffusion Neutronics," *Proceedings of the M & C 2015*.
- Talbot, P.W., K. Gamble, C. Rabiti, C. Wang, A. Alfonsi, J. J. Cogliati, D. Mandelli, and A. K. Prinja, 2016, "Time-Dependent Sensitivity Analysis of OECD Benchmark using BISON and RAVEN," *2016 ANS Winter Meeting and Nuclear Technology Expo*, Las Vegas, Nevada, November 6 through 10, 2016, submitted.
- Talbot, P.W., C. Wang, and C. Rabiti, 2016, "Multistep Input Reduction for High Dimensional Uncertainty Quantification," *Proceedings of the PHYSOR 2016*.



Talbot, P. W., A. K. Prinja, and C. Rabiti, 2016, “Adaptive Sparse-Grid Collocation Uncertainty Quantification Convergence for Multigroup Diffusion,” *Transactions of American Nuclear Society*.

Wang, C., P. W. Talbot, C. Rabiti, A. Alfonsi, D. Mandelli, and J. Cogliati, 2016, “An Efficient Sampling-Based Method for Sensitivity and Uncertainty Analysis Through RAVEN,” *Transactions of American Nuclear Society*.

### **Presentations**

Rabiti, C., J. Cogliati, G. Pastore, and A. Alfonsi, 2015, “Fuel Reliability Analysis Using BISON and RAVEN,” *International Topical Meeting on Probabilistic Safety Assessment and Analysis*, Sun Valley, Idaho, April 26 through 30, 2015.

Rabiti, C., P. W. Talbot, and C. Wang, 2015, “Development of tools and methodologies for uncertainty quantification and validation for the multi-physic fuel performance simulation,” DOE, Germantown, Maryland, September 22, 2015.

Rabiti, C., P. Talbot, A. Alfonsi, and G. Pastore, 2015, “Approaches for fast uncertainty quantification in continuous problems using RAVEN,” Ohio State University, May 2015, invited talk.

Talbot, P. W., C. Wang, and C. Rabiti, 2016, “Multistep Input Reduction for High Dimensional Uncertainty Quantification,” *PHYSOR 2016*, Sun Valley, Idaho, May 1 through 5, 2016.

Talbot, P.W., A. K. Prinja, and C. Rabiti, 2016, “Adaptive Sparse-Grid Collocation Uncertainty Quantification Convergence for Multigroup Diffusion,” *2016 ANS Annual Meeting*, New Orleans, Louisiana, June 12 through 16, 2016.

Wang, C., P. W. Talbot, C. Rabiti, A. Alfonsi, D. Mandelli, and J. Cogliati, 2016, “An Efficient Sampling-Based Method for Sensitivity and Uncertainty Analysis Through RAVEN,” *2016 ANS Annual Meeting*, New Orleans, Louisiana, June 12 through 16, 2016.

### **Invention Disclosures, Patents, Copyright**

The development described in the summary section has been implemented in the RAVEN code that is currently copyrighted to Battelle Energy Alliance, LLC.

### **Interns and Postdocs**

Interns: David B. Weitzel and Ana Jambrina Gomez

Postdoc: Congjian Wang

### **Collaborations**

University: University of New Mexico

## 14-098—Irradiation Effects of Uranium Dioxide

Jian Gan,<sup>1</sup> Lingfeng He,<sup>1</sup> Anter El-Azab,<sup>2</sup> and Sarah Khalil<sup>2</sup>

### General Project Description

Uranium dioxide (UO<sub>2</sub>) is the most widely used nuclear fuel in commercial light water reactors. Cumulative radiation damage during the fission process causes severe degradation in the thermophysical properties of UO<sub>2</sub> fuels, which limits their lifetime and increases their operational cost. Therefore, investigating defect production and evolution, fission product transport under irradiation, and revealing their physical mechanisms is of great importance in understanding degradation of the thermophysical properties of UO<sub>2</sub> fuels. In this work, both in situ and ex situ transmission electron microscopy (TEM) observation of defect nucleation and evolution under ion irradiation were conducted to understand radiation damage mechanisms in UO<sub>2</sub>. The irradiation-induced microstructure, including dislocation loops, inert gas bubbles, and lunar crater features are characterized using state-of-the-art characterization tools. In addition, the irradiation-induced microstructure in UO<sub>2</sub> has been directly compared to two uranium nitrides (i.e., UN and  $\alpha$ -U<sub>2</sub>N<sub>3</sub>).

### Summary

#### Experimental Effort

The microstructure evolution under high-dose irradiation in UO<sub>2</sub> has not been fully understood yet. The Intermediate Voltage Electron Microscopy-Tandem Facility at Argonne National Laboratory was employed to study defect evolution in situ in xenon (Xe)-irradiated UO<sub>2</sub>. The lunar crater features in UO<sub>2</sub> have been found for the first time. The thickness map measured by energy-filtered TEM shows the circular features having a crater shape (see Figure 1). In situ TEM observation shows that formation of crater features depends on the ion dose and irradiation temperature. The higher the irradiation temperature, the lower the dose at which the lunar crater features occur. The crater features occur at a dose of 315 dpa at 25°C, at a dose of 280 dpa at 600°C, and at a dose of 252 dpa at 800°C. An extensive study on chemical analysis using energy dispersive spectroscopy and electron energy loss spectroscopy indicates that the crater features have a similar composition to the matrix. High-resolution TEM results indicate they do not belong to any solid precipitates.

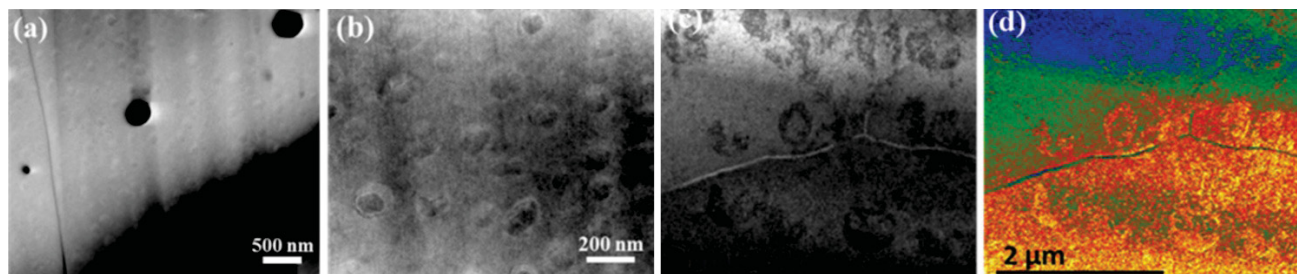


Figure 1. Lunar crater features in xenon-irradiated UO<sub>2</sub> at a dose of 350 dpa and 600 °C.

Xe-implantation introduced various types of defects such as interstitials and vacancies in UO<sub>2</sub>. Vacancies act as efficient trapping centers for the implanted Xe and turn it into Xe bubbles after trapping significant amounts of Xe. The implanted Xe gets segregated into bubbles in a molecular form, thereby creating internal pressure and taking them to an overpressurized state. When this internal pressure reaches the fracture limit of UO<sub>2</sub>, it ultimately lifts the implanted surface in the form of surface blistering/exfoliation. The bubble pressure at high

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<sup>2</sup> Purdue University

temperature is higher than that at low temperature, which makes the blistering/exfoliation occur at a lower dose. However, the detailed mechanisms of the surface blistering/exfoliation are very complicated and more effort is needed to understand them.

Proton irradiation on UN- $\text{UO}_2$  composites was carried out at the University of Wisconsin, which was supported early by a Nuclear Energy University Program project. Microstructure characterization of proton-irradiated composites has been partially supported by this project. High-resolution TEM and selected area electron diffraction images show that  $\alpha\text{-U}_2\text{N}_3$  and  $\text{UO}_2$  in UN/ $\text{UO}_2$  have the common axes  $[001]$ ,  $\alpha\text{-U}_2\text{N}_3 \parallel [001] \text{UO}_2$  and  $\alpha\text{-U}_2\text{N}_3[101] \parallel \text{UO}_2[101]$ . Special orientation relationships between  $\alpha\text{-U}_2\text{N}_3$  and  $\text{UO}_2$  may provide a better idea for designing a new nuclear fuel composite. The  $\alpha\text{-U}_2\text{N}_3/\text{UO}_2$  composites should be more stable than UN/ $\text{UO}_2$  composites. On the other hand, it also explains why some UN transformed to  $\alpha\text{-U}_2\text{N}_3$  when UN was sintered with  $\text{UO}_2$ .

$\text{UO}_2$  has an fcc structure of  $\text{CaF}_2$  type, UN has an fcc structure of NaCl type, and  $\alpha\text{-U}_2\text{N}_3$  has a bcc structure of  $\text{Mn}_2\text{O}_3$  type. The average dislocation loop size in  $\text{UO}_2$  (i.e., about 4 nm) is much smaller than those in nitride phases (i.e., 13 to 18 nm), which indicates that nucleation and growth of dislocation loops in  $\text{UO}_2$  is more difficult than in UN and  $\alpha\text{-U}_2\text{N}_3$ . Therefore, the radiation resistance of  $\text{UO}_2$  is better than UN and  $\alpha\text{-U}_2\text{N}_3$ . However, the effects on crystal structure, bonding strength, and oxygen or nitrogen vacancy on nucleation and growth of irradiation-induced defects are still not well understood. In addition, radiation effects on thermal transport of these three compounds need to be further investigated and compared in the future.

### ***Modeling Effort***

The goal of the modeling work was to use cluster dynamics to describe defect cluster population in irradiated  $\text{UO}_2$  and make a connection with previous work on extended x-ray absorption fine structure (EXAFS) characterization. During FY 2016, a previous analysis of cluster dynamics involving vacancy clusters has been advanced to only include interstitial clusters. Including interstitial data in the analysis enabled us to apply cluster dynamics to the void and loop formation in irradiated  $\text{UO}_2$  and focus on the oxidation state of the matrix to make the connection with EXAFS. By lumping all defect clusters above monomers together in stoichiometry calculations and assuming that mono defects determine the off-stoichiometry of the matrix, it was found that (a) the fraction of off-stoichiometric clusters is higher than the fraction of stoichiometric ones; (b) hypo-stoichiometric clusters dominate at low temperature while hyper-stoichiometric clusters dominate at high temperatures; (c) the rate of absorption of oxygen vacancies into clusters is faster than that of uranium vacancies, which leads to growth of oxygen-vacancy rich voids (these voids are called hyper-stoichiometric); and (d) the  $\text{UO}_2$  matrix tends to oxidize under irradiation. Uranium vacancies have the highest concentration amongst other monomers in the matrix, which means the matrix is hyper-stoichiometric mainly due to accumulation of uranium vacancies.

### **Benefits to DOE**

This project was the first time for creating lunar crater features in  $\text{UO}_2$  through ion irradiation. In situ studies of crater feature formation in  $\text{UO}_2$  using the Intermediate Voltage Electron Microscopy -Tandem Facility provides a unique opportunity for studying the effects of bubble pressure on microcracking, blistering, and exfoliation in  $\text{UO}_2$  and other materials. Direct comparison of radiation-induced microstructure among  $\text{UO}_2$ , UN, and  $\text{U}_2\text{N}_3$  shows better radiation damage tolerance of  $\text{UO}_2$ . INL has benefited from this work by further enhancing its research and development strength on fuel material microstructure characterization and guiding the design of next generation nuclear fuels. Our work has also contributed to DOE's leading role on basic science research of materials behavior and performance in extreme environments.

## Publications

- He, L., J. Gan, and T. R. Allen, 2016, “Microstructure Evolution in Ion-Irradiated  $\text{UO}_2$ ,” *Transactions of the American Nuclear Society*, in press
- He, L., J. Pakarinen, B. Jaques, A. T. Nelson, J. Gan, A. El-Azab, D. Butt, and T. R. Allen, “Bubble Evolution in Kr Ion Irradiated  $\text{UO}_2$  at High Temperature,” to be submitted.
- He, L., J. Gan, and M. Kirk, “Lunar crater features in  $\text{UO}_2$ ,” to be submitted.
- He, L., B. Tyburska-Pueschel, B. Jaques, and J. Gan, “Radiation damage on  $\text{UO}_2$  and UN,” to be submitted.
- Khalil, S., T. Allen, and A. El-Azab, “Off-Stoichiometric Cluster Dynamics Modeling of  $\text{UO}_2$ ,” in preparation for submission to *Journal of Chemical Physics*.
- Khalil, S., T. Allen, and A. El-Azab, “Off-Stoichiometric Defect Clustering in Irradiated Oxides,” to be submitted to *Materials Science Journal*.

## Presentations

- He, L., J. Gan, M. Kirk, B. Tyburska-Pueschel, and B. Jaques, 2017, “Radiation Damage on  $\text{UO}_2$  and UN,” *TMS 2017 Annual Meeting and Exhibition*, San Diego, California, February 26 through March 2, 2017, invited.
- He, L., J. Gan, and T. R. Allen, 2016, “Microstructure Evolution in Ion-Irradiated  $\text{UO}_2$ ,” *American Nuclear Society Annual Meeting 2016*, New Orleans, Louisiana, June 12 through 16, 2016, poster.
- He, L. and J. Gan, 2016, “Characterization of Radiation-Induced Microstructure in  $\text{UO}_2$ ,” *Advances in Structural and Chemical Imaging (ASCI) Symposium*, Boise, Idaho, May 18 through 20, 2016, invited.
- Khalil, S., A. El-Azab, T. Allen, and A. Hamed, “Cluster Dynamics in Irradiated  $\text{UO}_2$ : Off-Stoichiometric Considerations for Voids, Loops and the Oxide Matrix,” Symposium ZZ—Material Design and Discovery via Multi-Scale Computational Materials Science, *MRS Fall 2015 Meeting*, Boston, Massachusetts, November 29 through December 4, 2015.
- Khalil, S., T. Allen, and A. El-Azab, 2016, “Cluster Dynamics Modeling of Extended Defects in Irradiated  $\text{UO}_2$  with Off-Stoichiometry Considerations,” *TMS 2016 145th Annual Meeting & Exhibition*, Nashville, Tennessee, February 14 through 18, 2016.

## Collaborations

University: Illinois Institute of Technology, University of Wisconsin, and Purdue University

## 14-104—Development of a Multiphysics Algorithm for Analyzing the Integrity of Nuclear Reactor Containment Vessels Subjected to Extreme Thermal and Overpressure-Loading Conditions

Richard C. Martineau,<sup>1</sup> Ben Spencer,<sup>1</sup> Ray Berry,<sup>1</sup> David Andrs,<sup>1</sup> Hongbin Zhang,<sup>1</sup> and Yidong Xia<sup>1</sup>

### General Project Description

The technical objective of this project is to develop a first-of-a-kind, multi-scale, multi-physics algorithm for analyzing the integrity of nuclear reactor containment vessels subjected to extreme thermal and overpressure loading conditions. The various specific physical phenomena to be considered in developing an algorithm for analyzing nuclear containment events are as follows:

1. Single-phase, multi-component, and multi-phase flow and transport
2. Chemical reactions, including combustion and deflagration to detonation transition
3. Shock wave formation and propagation
4. Conjugate heat transfer, including solid-state and hydrodynamic heat conduction, forced and natural convection, and radiative heat transfer
5. Fluid-structure interaction (thermo-mechanical-hydrodynamic coupling).

INL's Multi-Physics Object-Oriented Simulation Environment (MOOSE) high-performance computing development and runtime computational framework will be used for this research effort. The central focus of the algorithmic research and development for this effort will take place in Bighorn (i.e., the MOOSE-based application for single and multi-phase conjugate heat transfer). The MOOSE software repository contains some of the necessary physics modules (denoted as kernels) that are readily available for this effort. Because of the MOOSE framework's unique modular design, these kernels may be leveraged with little or no modification of the source code. Additional kernel development will be required for gas-phase chemical reactions, shock wave capture, fluid-structure interaction, and thermal radiation. Integrating these physics into a single strongly coupled multi-physics algorithm is the research task at hand. The benefit of this algorithmic development will be delivery of a multi-dimensional MOOSE-based application capable of analyzing the integrity of nuclear reactor containment vessels subjected to extreme thermal and overpressure-loading conditions.

### Summary

#### ***Radiative Heat Transfer***

Rattlesnake is a MOOSE-based application initially designed as a neutron transport solver. Various discretization schemes have been implemented in Rattlesnake for solving the multi-group neutron transport equation. The neutron transport equation and the radiation transport equation (RTE) share some similarities and also differ from each other. They both have streaming direction as an independent variable. RTE has the similar time-derivative kernel, streaming kernel, collision kernel, and scattering kernel. (The name 'kernel' is borrowed from the MOOSE framework, in which a kernel stands for a piece in the weak form for a particular term in the equation.) The new kernels introduced in RTE include the black-body emission kernel and the spatial varying refractive index kernels. We frequently solve the linear eigenvalue problem for neutron transport. On the other hand, RTE is typically coupled with the energy transfer equation in a strong nonlinear fashion. Nonlinear transient solvers are required for solving RTE. Outputs for modeling radiative heat transfer can be maximized by using

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<sup>1</sup> Idaho National Laboratory

Rattlesnake/MOOSE with minimum effort. Rattlesnake was extended for radiative heat transfer with all discretization schemes, solvers, and so on being available for solving the multi-group RTE.

### ***Multi-Physics with MOOSE Multi-Apps and Transfers***

In order to improve full core simulation fidelity, several enhancements were added to the MOOSE framework for this LDRD effort and these improvements include improved “Multi-app” integration, more robust “transfers,” new transfer types, and refactored stepping and time control. MOOSE contains multi-apps and transfers, which are features in MOOSE that allow solution transfers between an arbitrary number of applications. The multi-app system was redesigned to better support multi-level simulations with support for restart and recovery for long-running simulations. The new system is now capable of saving checkpoints throughout the multi-app hierarchy, which can restore the state of a simulation up to a valid state due to planned or unplanned termination.

### ***Computational Fluid Dynamics and Shock Capturing***

The Bighorn computational fluid dynamics MOOSE-based application now has the algorithmic capability for modeling and predicting the multi-component, multi-phase fluid dynamics in multi-dimensions. Bighorn has continuous finite element and discontinuous Galerkin spatial solution of a coupled system of fluid flow and heat transfer. Although multi-fluid is not fully developed, Bighorn has been designed to handle multi-fluid problems. Because of the flexibility in the underlying MOOSE framework, Bighorn is quite extensible and can accommodate both multi-species and multi-phase formulations.

### **Benefits to DOE**

This work is directed toward creating a new algorithm for thermal-mechanical fluid-structure interaction to address structural integrity in the presence of thermal radiation, natural and forced convection, and strong hydrodynamic forces. While this effort is directed with nuclear safety for nuclear power plants in mind, applications for nuclear security are numerous.

### **Publications**

- Xia, Y., D. Andrs, and R. C. Martineau, 2016, “BIGHORN Computational Fluid Dynamics Theory, Methodology, and Code Verification & Validation Benchmark Problems,” Technical Report, INL/EXT-16-39214, Idaho National Laboratory, Idaho Falls, Idaho, August 2016.
- Xia, Y., D. Andrs, and R. C. Martineau, 2016, “A Hermite WENO-based Slope Limiter for Finite Volume Method on Unstructured Grids,” INL/JOU-16-39840, *Journal of Computational Physics*, in preparation.

### ***Presentations***

- Xia, Y. and R. C. Martineau, 2016, “Development of a Multiphysics Algorithm for Analyzing the Integrity of Nuclear Reactor Containment Vessels Subjected to Extreme Thermal and Overpressure-Loading Conditions,” INL/CON-16-39628, presented at INL Annual LDRD Poster Exhibition, Idaho Falls, Idaho, August 2016.



## 15-002—Experimental Scenarios of Adversity and Recovery in Aqueous Separations

Peter Zalupski<sup>1</sup> and Travis Grimes<sup>1</sup>

### General Project Description

The AKUFVE apparatus is an instrument for rapid and accurate measurements of distribution coefficients, designed with a specific aim of streamlining the ability to continuously monitor movement of materials in re-circulating immiscible liquid phases. This flow system can be directly connected to various detectors, enabling radiometric, spectrophotometric, atomic absorption, pH, and temperature measurements, while liquid delivery is supported by attached burettes. This configuration offers a unique opportunity for dynamically studying aqueous separations as conditions that are intentionally varied to monitor either beneficial or detrimental effects. The main objective of this research project is incorporation of online monitoring tools around the AKUFVE recirculating unit. Demonstration of this experimental capability may establish new collaborative relationships in the field of recovery for valuable materials from a variety of hydrometallurgic streams.

### Summary

In FY 2015, this mixer-centrifugal unit was installed in Laboratory B-209 at the Energy Innovation Laboratory building. Experimental testing in FY 2016 focused on the operational capability of the AKUFVE. Successful demonstration of its operation allowed efficient circulation of two immiscible liquid phases: (1) acidic water and (2) lamp oil, between the mixing chamber (where two phases are equilibrated) and the H-centrifuge (where two phases are cleanly separated). Figure 1 shows operation of the unit, where two immiscible phases are mixed and two clean phases are being recirculated into the mixing chamber after H-centrifuge separation. At this particular stage, as the separated liquids return to the mixing chamber, the liquid-liquid distribution equilibrium may be perturbed to yield a vast spectrum of information in search of efficient methodologies for recovery of valuable resources via hydrometallurgical means. This is a unique experimental capability that focuses on studying equilibrium changes using detectors installed inline and performing measurements on clear, separated phases returning to the mixing stage.

During FY 2016, the spectrophotometric measuring capability was being incorporated. Two custom-made, fiber optic T-flow-through cells (custom sensors and technology) are included in the light and heavy-phase recirculating loops, allowing monitoring of light absorption in the ultraviolet-visible wavelength range using two QEPro spectrophotometers (Ocean Optics, Inc.). The operation of the spectrophotometers was validated through monitoring spectral changes resulting from the complexation of neodymium ion by an aminopolycarboxylate reagent. Figure 2 illustrates the experimental data collected, along with the calculated molar absorptivities of two light-absorbing species in solution. The refinement of experimental data produced well-defined thermodynamic stability constants for the monitored complexation reactions.

Also during FY 2016, two new laboratory hoods were installed in Laboratory B-209 to accommodate operation of AKUFVE unit.

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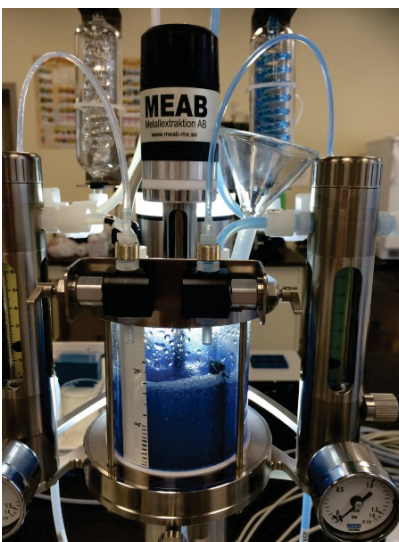


Figure 1. Lamp oil/acidic water experiment in AKUFVE.

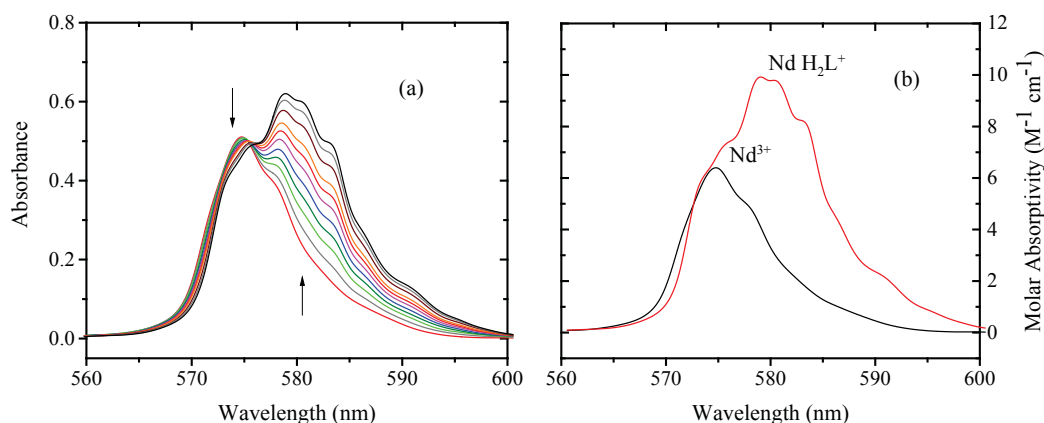


Figure 2. Spectrophotometric validation of QEPro spectrophotometers using the  $\text{Nd}^{3+}$ /aminopolycarboxylic acid complexation reaction.

## Benefits to DOE

Demonstration of this new online monitoring tool for tracking movements of valuable materials in mixtures designed for their recovery and purification may attract multiple new customers and collaborative relations. A dynamic means of monitoring liquid-liquid partitioning equilibria streamlines experimental research and a variety of scenarios may be simulated to showcase the AKUFVE design. Demonstrations will target interest in this experimental capability from a variety of partners, both domestic and international. Potential sources of follow-on funding could include the Fuel Cycle Research and Development program (An/Ln), Critical Materials Institute (rare earths), Environmental Management (remediation), the National Nuclear Security Administration (highly efficient purification), and international partnerships.

## Interns and Postdocs

Postdoc: Colt Heathman

## 15-013—Simulation-Based Analysis of Procedures and Accident Management Guidelines

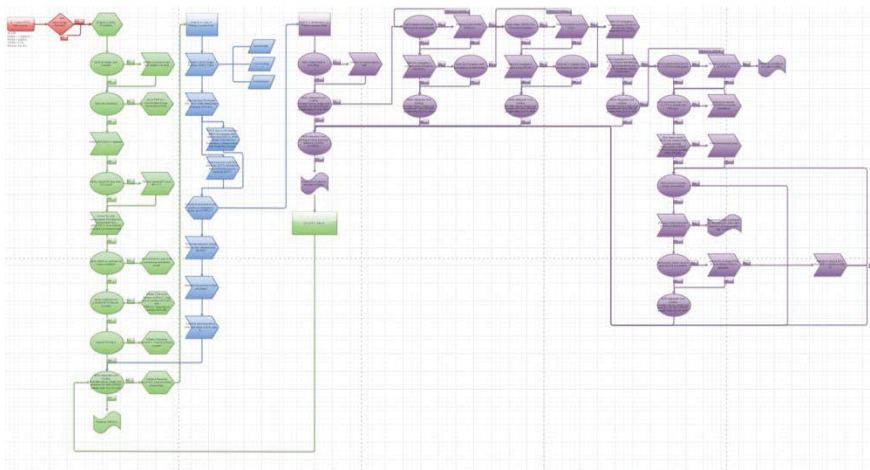
Curtis Smith,<sup>1</sup> Tony Koonce,<sup>1</sup> Ronald Boring,<sup>1</sup> and Robert Youngblood<sup>1</sup>

### General Project Description

The objective of this project is to improve the state-of-the-art of probabilistic risk analysis (PRA) by modeling plant logic, procedures, and accident management guidelines in a unified way. Existing fault-tree/event-tree risk models address human performance in a largely ad hoc way; incorporating a complete set of procedures and guidelines into a simulation model of plant risk is well beyond the state-of-practice. The research aims to advance the technical basis for enabling implementation of simulation-driven predictive accident guidelines as a means of addressing uncertainty in the selection of actions by projecting the effect of those actions into the future. Toward this goal, the project is investigating the feasibility of a computerized online decision support system for risk-informed management. Using simulation of procedures, we will be forecasting decision-support options (for plant operators) from a large spectrum of simulation-based scenarios. The novel aspect of the work is inclusion of a comprehensive set of procedures and accident management guidelines in a simulation model of plant performance. This is technically important because, despite decades of work on human reliability methods for PRA, procedurally mandated actions still surprise observers (e.g., shutting off high-pressure injection at Three Mile Island or shutting off systems at Fukushima), partly because of the ad hoc and generally incomplete treatment of human performance in state-of-practice PRA.

### Summary

Research has proceeded to the point of investigating a computerized online decision support system for risk-informed management of beyond design basis accidents. Using RAVEN, we are forecasting decision support options (for plant operators) from a large spectrum of simulation-based scenarios. The novel aspect of this work is inclusion of a comprehensive set of procedures and accident management guidelines in a simulation model of plant performance. The LDRD team reviewed the various events and emergency procedures recommended for the ATR. The team decided to pursue the loss of primary coolant flow initiating event and its impacts on E-0, Entry Procedure, and transition into subsequent emergency procedures. The team used this event to develop the simulation methodology that will be used throughout this project.



Our simulation methodology was designed to resemble an operating nuclear plant by actively modeling the various attributes that constitute the as-built and as-operated plant. The as-built aspect of the plant is included in our simulation by incorporating the plant's thermal-hydraulic model, equipment reliability data for plant components, and plant indications and alarms. The simulation code developed during FY 2016 is currently being written in the language Python to represent a

complex ATR-based procedure. This simulation serves as the interface between the procedures and plant response

<sup>1</sup> Idaho National Laboratory

(i.e., in RELAP-5, the current thermal hydraulics code for ATR). An example of procedure steps is shown in the figure on the previous page.

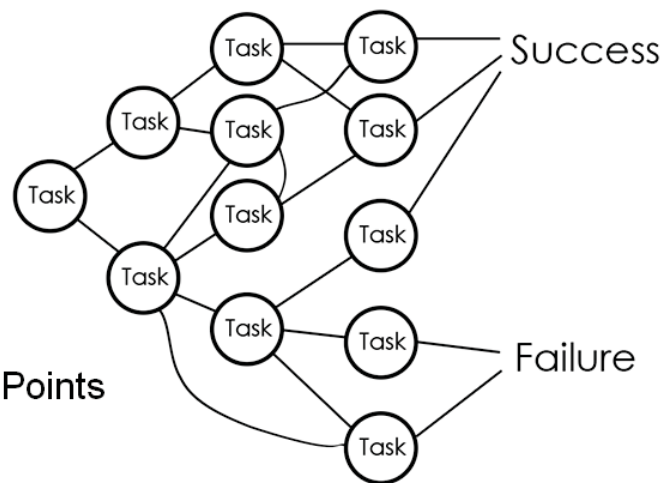
The team has developed an initial simulation approach to represent operator actions while implementing mitigating procedures for ATR initiating events. The team continues to develop the simulation code; code libraries continue to be populated with plant procedures, components, indications, and operator actions. RELAP-5 is the selected thermal-hydraulics software for this project in order to take advantage of current ATR modeling done in this code. Modeling work will also incorporate a portion of the HUNTER human reliability analysis framework (the “primitives” that represent individual operator actions or steps) to build operator error events and probabilities within procedure simulation. The simulation is focused on tasks within procedures (see figure at right) that should be applicable to HUNTER information.

### Benefits to DOE

The capability to be developed has the potential to materially improve the state-of-practice of PRA, affecting facilities at INL and

elsewhere and promoting work for INL staff. INL will benefit by the latest developments in PRA that may also be applied to DOE facilities. The research is planned to be performed using ATR as the case study. Upon proof of principle, the technique could be used by both the nuclear industry and regulator, specifically including the Light Water Reactor Sustainability Program. In addition, there is some potential for expanding this methodology beyond the nuclear industry and adapting it to other low probability/high consequence industries and infrastructure.

### Simulation Points



## 15-023—Development of Stochastic Three-Dimensional Soil Response Capability in MOOSE to Provide Design and Beyond Design Basis Seismic Motions for Nuclear Facilities

Justin L. Coleman<sup>1</sup> and Swetha Veeraraghavan<sup>1</sup>

### General Project Description

Soil material properties at every site have large variability. The stochastic nature of these soil material properties is currently modeled in the industry using an indirect approach involving a statistically significant number of deterministic analyses. A more theoretically accurate approach is to perform stochastic site-response analyses, which will be used in this project after developing a stochastic elasto-plastic finite element formulation in MASTODON, which is a Multi-Physics Object-Oriented Simulation Environment (MOOSE) framework application. Understanding the site response is significant because it has a direct effect on the energy transferred to a structure during an earthquake.

This project will provide two new numerical capabilities in the MOOSE-based application, MASTODON:

1. Three-dimensional seismic wave propagation in time domain computations
2. Stochastic finite elements (allows for direct computation of variability in soil material properties).

### Summary

This project aims at developing two new capabilities in the MOOSE-based application MASTODON to more realistically simulate seismic response of nuclear facilities to earthquake excitation. The following capabilities have been added to MASTODON in the past year to enable the calculation of three-dimensional seismic wave propagation in time domain:

- In reality, soil domains are infinite. When a soil domain is truncated for a computer simulation, waves get reflected off these truncated boundaries. Absorbing boundaries have been implemented to absorb the waves incident on the truncated boundaries to simulate wave propagation in an infinite soil domain.
- Any earthquake fault ruptures can be modeled using a series of point sources. Each of these point sources releases a part of the seismic energy. The capability of applying one or multiple point sources to simulate any earthquake fault rupture has been included in MASTODON.
- A domain reduction method has also been implemented in MASTODON. This method is particularly useful when a spatially big domain (in the order of many kilometers) has to be re-run multiple times due to changes that are localized in a small region (such as different structural parameters localized to a domain of few meters). This method was implemented in MASTODON with the help of Professor Jacobo Bielak (Carnegie Mellon University).
- A suite of verification problems have been developed and a comparison between results obtained from MASTODON and other commercial software such as LS-DYNA (LSTC 2013) and DEEPSOIL (Hashash et al. 2011) is within acceptable tolerances. Verification was conducted in collaboration with Professor Youssef Hashash and his students (Omar Baltaji and Ozgun Numanoglu).

The capabilities mentioned above have been used to simulate the response of a nuclear containment building and reactor pressure vessel to earthquake ground motion from an inclined fault rupture. This response of the pressure

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vessel was coupled with a fuel rod model created in BISON to simulate the thermal and mechanical response of the fuel rod when a scram is triggered due to the earthquake.

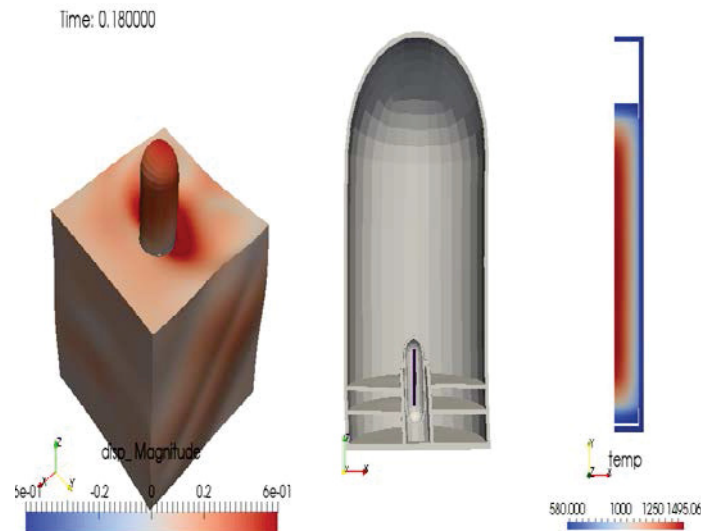


Figure 1. Coupled MASTODON/BISON simulation. The image to the left shows the soil domain with the inclined earthquake waves. The center image shows the response of the containment building, pressure vessel, and a fuel rod inside the pressure vessel. The image to the right shows the temperature profile of the smeared fuel pellets and the clad when the scram is triggered.

**Ongoing work:** There are many uncertainties when it comes to modeling the response of soils to earthquake excitation. Variability in soil properties can be fit with a probability distribution. When one value is chosen for a soil parameter, only one realization of the problem is simulated. Stochastic methods have to be employed to evaluate the effect of uncertainty in the input (i.e., soil parameters) on the final quantity of interest (e.g., acceleration at a location inside the reactor). Two ways to accomplish this are as follows:

- Monte Carlo is a commonly used calculation. In this technique, multiple realizations of the problem are simulated and the output is obtained for each realization. This output from the multiple realizations can then be combined to obtain the probability density function, which characterizes the uncertainty in the output. Currently, RAVEN and MASTODON are being coupled to add this capability of automatically simulating multiple realizations of the problem to obtain uncertainty in the output.
- Stochastic finite element method is used in other computation applications, but it is new to seismic simulations in soils. Another method of estimating the uncertainty in the output is by introducing this quantity as a variable into the problem. In this method, the output uncertainty is automatically calculated by running just one simulation instead of the multiple simulations required in the Monte-Carlo method. Efforts are underway to add this capability into MASTODON.

## Benefits to DOE

Development of a stochastic site response capability as outlined in this report provides an advanced tool for stochastically simulating the propagation of seismic waves and evaluating seismic risk at nuclear facilities. Existing tools may cause nuclear facility owners to spend more money than necessary to mitigate perceived seismic risk. The new capability provided by this project has potential to remove additional conservatism from the design and analysis of existing and new nuclear facilities. This removal of conservatism translates to dollars.



## References

Hashash, Y. M. A., D. R. Groholski, C. A. Phillips, D. Park, and M. Musgrove, 2011, "DEEPSOIL 5.0 User Manual and Tutorial," 107 p.

LSTC, 2013, "LS-DYNA Keyword User's Manual - Version R 7.0," Livermore Software Technology Corporation, Livermore, California.

## Publications

Journal article in progress, abstracts submitted for SMiRT 2018 conference papers and presentations

### ***Presentations***

Coleman, J. and S. Veeraraghavan, 2016, "Seismic Research and Development Initiative," DOE Headquarters to DOE-NE 2, February 24, 2016.

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Veeraraghavan, S. and J. Coleman, 2016, "Development of MASTODON and Applications," *DOE Natural Phenomena and Hazards DOE Headquarters Working Meeting*, October 18, 2016.

Veeraraghavan, S., J. Coleman, and B. Spencer, 2016, "Effect of Soil Heterogeneity on Nuclear Facility Soil Foundation Interaction," *Engineering Mechanics Institute Conference 2016*, Nashville, Tennessee, May 2016.

## Interns and Postdocs

Interns: Ozgun Numanoglu, Omar Baltaji, and Andrea Castellano

Postdoc: Swetha Veeraraghavan

## Collaborations

University: University of California, Davis

## 15-032—Development of New Method for High-Temperature Thermal Conductivity Measurements of Nuclear Materials

Krzysztof Gofryk<sup>1</sup> and Robert Mariani<sup>1</sup>

### General Project Description

The thermal conductivity of nuclear fuels governs the conversion of heat produced from fission events into electricity and is an important parameter in reactor design and safety. Different scattering mechanisms affect thermal conductivity at different temperatures. Therefore, in order to fully understand and reliably model this property, it is crucial to study thermal conductivity in a broad temperature range. However, blackbody infrared radiation makes this task challenging above room temperature and several transient techniques have been developed to overcome this issue. Our objective is to extend measurement capability of our new state-of-the-art PPMS experimental setup from 350 to 800 K using the so-called  $3\omega$  (omega) method. This *ac* technique is considered to be the best pseudo-contact method available and enables direct measurement of thermal conductivity of very small samples. This is rare for transient thermal transport measurements because independent determinations of the thermal diffusivity and heat capacity are required to derive the thermal conductivity. In addition, this technique intrinsically has the lowest possible errors caused by infrared radiation, and the error-term arising from the radiation is estimated to be less than 2%, even at 1000 K. Adaptation of this method will be novel for nuclear materials. The new experimental setup will allow study of the thermal conductivity of small samples of nuclear materials in an exceptionally broad temperature range (2 to 800 K), and the data obtained will help to advance the theoretical understanding and modeling of reactor fuel and future fuel development.

### Summary

The goal of the project is to develop a new method for thermal conductivity measurements of nuclear materials using the  $3\omega$  method.

During the second year of the project, we (1) designed and built an experimental setup for thermal conductivity measurements of metallic materials. This setup constitutes the high-resolution lock-in amplifier, high precision voltmeter, and programmable current source. All devices are controlled or developed by computer software based on the LabView platform. We also (2) purchased an ARS cryocooler for thermal conductivity measurements as a function of temperature in the range of 5 to 800 K; (3) purchased a metal sputter coater and installed a new optical microscope; and (4) performed high-resolution thermal conductivity measurements of selected reference metallic samples (Pt, Cu, Alumel) using  $3\omega$ -method. Figure 1 shows an example of thermal conductivity determination of Pt wire using the new method. The results have been compared to reference data (see Figure 2).

### Benefits to DOE

Thermal conductivity is a key engineering parameter in the conversion of heat produced by fission events to electricity. Therefore, new reliable methods for thermal conductivity determinations of nuclear materials, such as the  $3\omega$  technique, along with advanced modeling are key parameters in understanding and developing new nuclear fuels, leading to more efficient energy production and security of the nation. While developed for small bulk samples, this method may also be applied for micro-sized and thin-film materials, which are unique and can be extended in the future to active materials such as neptunium, plutonium, and americium-based phases. In addition, this *ac* thermal conductivity method could be modified to measure *ac* Seebeck coefficient ( $2\omega$ -method) and heat capacity. Altogether, this cutting edge research will help INL to maintain a leading position in nuclear fuel research.

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<sup>1</sup> Idaho National Laboratory

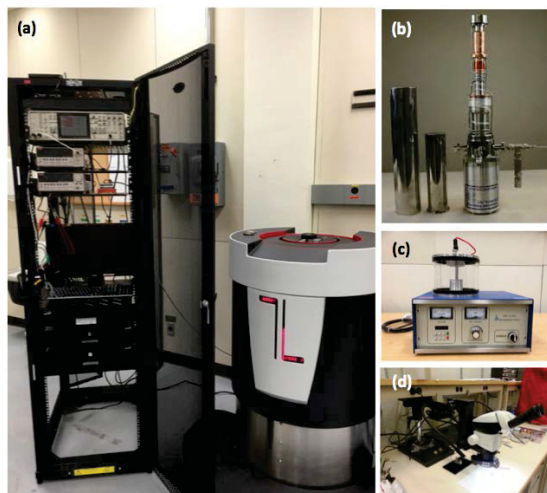


Figure 1. The main equipment that had been purchased for  $3\omega$  development and installed in IF-603, Laboratory C6. (a) Experimental setup, along with a PC (all controlled by LabView software); (b,c,d) ARS cryocooler, metal sputter, and microscope, respectively.

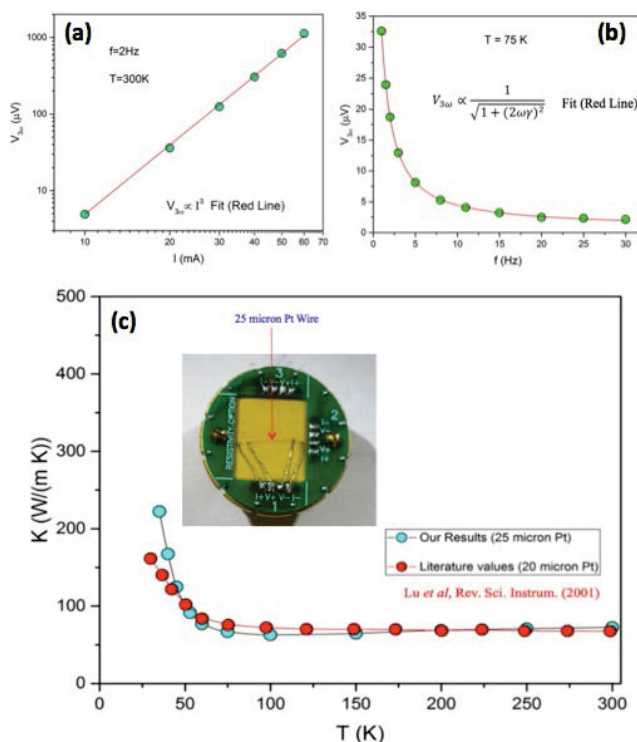


Figure 2. Example of thermal conductivity measurements of Pt wire using  $3\omega$  technique. (a,b)  $3\omega$  signal as a function of ac current and frequency and (c) the thermal conductivity of Pt wire measured by our  $3\omega$  setup. Inset: Pt wire mounted on PPMS puck and ready for testing.

## **Publications**

Two papers are currently in preparation and should be completed by the end of FY 2017.

## **Interns and Postdocs**

Postdocs: Keshave Shrestha and Daniel Antonio

## 15-040—Acoustic Telemetry Infrastructure for In-Pile ATR and TREAT Monitoring

Vivek Agarwal,<sup>1</sup> James A. Smith,<sup>1</sup> James Lee,<sup>1</sup> and James K. Jewell<sup>1</sup>

### General Project Description

The research project will develop an acoustic measurement infrastructure (AMI) to obtain an acoustic baseline of the ATR under different operating conditions. This presents an opportunity to (1) identify quiescent frequency ranges that would aid in design of the carrier frequency of acoustically telemetered sensors, (2) enhance/tune the acoustically telemetered sensor design prior to fabrication and installation inside the reactor core; and (3) understand and characterize the salient measurement parameters of an actual thermoacoustic sensor signal: frequency, amplitude, and phase. AMI can be adapted for Transient Reactor Test Facility. Current implementation of AMI includes (1) ATR; (2) acoustic sources from primary coolant pumps (PCPs); (3) acoustic transducers installed on the surface of the reactor vessel; and (4) signal processing algorithms to develop the first-ever ATR acoustic baseline signatures (Figures 1 and 2).

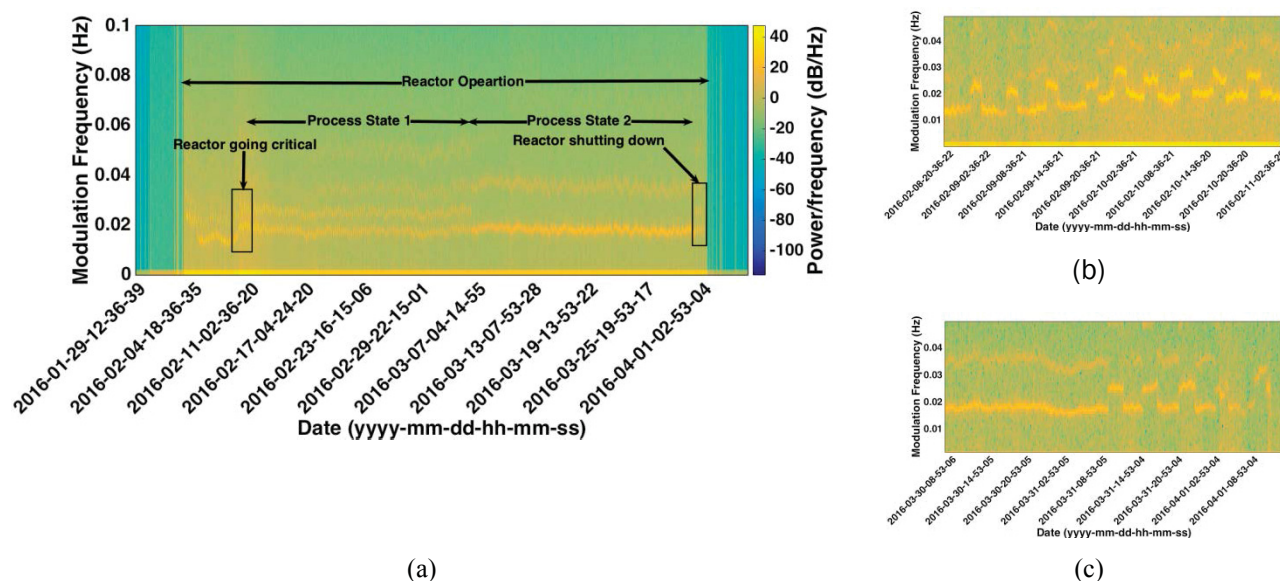


Figure 1. (a) Acoustic baseline signature of ATR recorded during reactor operation using AMI, (b) zoomed in view of modulation frequency as reactor approaches and achieves criticality, and (c) zoomed in view of modulation frequency variations observed before reactor shutdown occurs.

### Summary

The fundamental innovative premise behind AMI is the belief that listening to a reactor's intrinsic and extrinsic acoustic sensors will enable (1) real-time non-intrusive in-pile measurement that would aid development and validation of computational models and micro-scale understanding of nuclear fuel and materials; (2) wireless (i.e., cable-less) communication of key in-pile parameters through reactor structures like flanges, pressure vessel

<sup>1</sup> Idaho National Laboratory

body, and pneumatic tube; and (3) self-powered acoustically telemetered sensor for in-pile sensing during irradiation.

It has been established that frequency harmonics generated by PCP vanes are nearly identical to signals that would be generated by acoustically telemetered sensors like the thermoacoustic sensor [1] when inserted into the reactor core. Therefore, periodic pressure pulses from a PCP make an excellent “virtual” acoustically telemetered signal and are used for developing ATR acoustic signatures.

ATR acoustic baseline signatures collected during atypical and Powered Axial Locator Mechanism operational cycles using AMI are shown in Figures 1 and 2, respectively. During a typical ATR operational cycle, two PCPs are operated and reactor power is at a nominal level. During a Powered Axial Locator Mechanism operational cycle, three PCPs are operated and reactor power is increased above the nominal level. In Figure 2, two amplitude modulations are observed (bolded yellow line) – one between PCPs 6 and 9 (near 0.05 Hz) and another one between PCPs 6 and 8 (near 0.025 Hz). However, during a typical operational cycle, one amplitude modulation near 0.025 Hz is observed. Figures 1 and 2, highlights changes in the ATR acoustic baselines under different operating conditions.

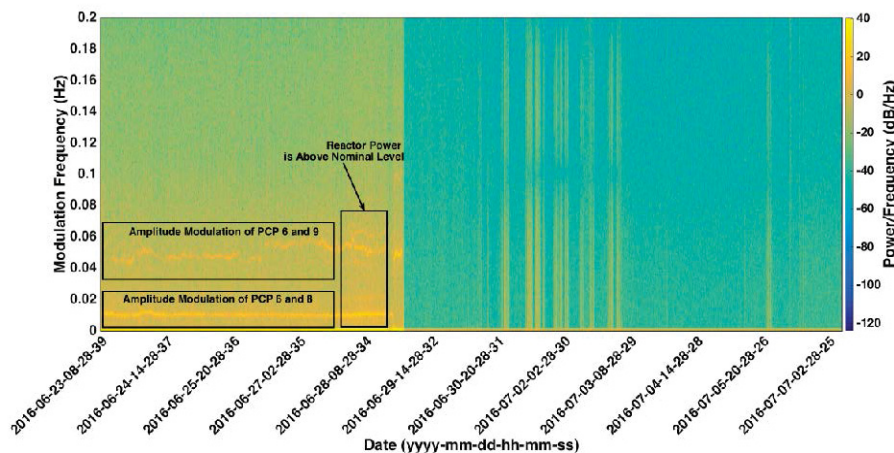


Figure 2. Acoustic baseline of ATR recorded during Powered Axial Locator Mechanism cycle at a flange.

## Benefits to DOE

This research is critical to advancing DOE’s nuclear energy mission by developing an advanced capability of in-pile temperature measurements for materials irradiation in research and test reactors. This aligns with the Nuclear Science User Facilities mission in the area of advanced instrumentation for in-pile measurements. The research will enable direct technology and capability transfer opportunity to the nuclear industry to support current and next generation reactor concepts. Successful completion of this research will significantly improve the quality and capability of INL to analyze in-pile data for materials, fuels, and carry out modeling and simulation research and development. This project has also allowed researchers to work closely with ATR personnel. ATR turned to AMI to supply vibration data to help understand the beryllium cracking issue near the hydraulic shuttle. The research augments the activities outline within the INL’s Nuclear Science and Technology Directorate’s Gateway to Accelerated Innovation in Nuclear Initiative.

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- [1] Ali, R. A., S. L. Garrett, J. A. Smith, and D. K. Kotter, 2013, “Thermoacoustic Thermometry for Nuclear Reactor Monitoring,” *IEEE Journal of Instrumentation and Measurement*, Vol. 16, No. 3, pp. 18–25.



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- Agarwal, V. and J. A. Smith, 2017, “Real-Time In-Pile Acoustic Measurement Infrastructure at the Advanced Test Reactor,” accepted *ANS Nuclear Technology* (to appear in March 2017).
- Hrisko, J., S. L. Garrett, W. M. Smith, J. A. Smith, and V. Agarwal, 2015, “The Vibroacoustic Environment in a Nuclear Reactor,” *169<sup>th</sup> Meeting of Acoustical Society of America*, Pittsburgh, Pennsylvania, May 15 through 22, 2015.
- Smith, J. A., B. J. Heidrich, V. Agarwal, M. D. Heibel, and S. L. Garrett, 2016, “Using the Sounds of Nuclear Power,” *171st Meeting of the Acoustical Society of America*, Salt Lake, Utah, May 23 through 27, 2016, (presented in a Special Invited Session on Thermal Acoustics sponsored by three ASA Technical Committees: Structural Acoustics, Vibration and Engineering Acoustics, and Physical Acoustics).
- Smith, J. A., S. L. Garrett, M. D. Heibel, V. Agarwal, and B. J. Heidrich, 2016, “Progress In Developing An In-Pile Acoustically Telemetered Sensor Infrastructure,” *American Nuclear Society TOPFUEL*, Boise, Idaho, September 11 through 16, 2016.
- Smith, J. A., S. L. Garrett, M. D. Heibel, and V. Agarwal, 2016, “Wireless In-Core Acoustic Telemetry and Self-Powered Sensing,” *Nuclear Energy Enabling Technologies Crosscutting Technology Development Advanced Sensors and Instrumentation Newsletter*, Issue 3, February 2016.

## Presentations

- Agarwal, V., 2016, “Acoustic Measurement for In-Pile ATR and TREAT Monitoring – Using the Sound of Nuclear Power,” DOE Idaho Operations Office, September 7, 2016, Idaho Falls, Idaho.

## Invention Disclosures, Patents, Copyrights

- IDR No. BA-893, Tracking No. 4112, “Acoustic Measurement Infrastructure Method and System for Process Monitoring, Diagnostics, and Prognostics,” provisional patent filed.

## Interns and Postdocs

Interns: Alexander Pharr and Joshua Hrisko

## 15-060—Development of Efficient TREAT Modeling Capabilities with Graphite Data Improvement

Mark DeHart,<sup>1</sup> Ayman Hawari,<sup>2</sup> Todd Palmer,<sup>3</sup> Edward Blandford,<sup>4</sup> and Benoit Forget<sup>1</sup>

### General Project Description

This project consists of a collection of independent research from university researchers under the INL National University Consortium. The original proposal for this project called for a collaborative effort between the Massachusetts Institute of Technology (MIT) and the University of New Mexico (UNM). However, the National University Consortium Program revised the scope and participants, resulting in four separate research projects that were coordinating only with INL. Although independent, the research efforts all support modeling and experimentation related to the Transient Test Reactor (TREAT). Ayman Hawari of North Carolina State University (NCSU) is performing modeling of the TREAT M2 calibration series core to support validation of INL tools; Wade Marcum of Oregon State University (OSU) supported research related to phonon transport in graphite using the INL Rattlesnake application during his first year, Todd Palmer lead research to develop a benchmark of the TREAT minimum core configuration, followed by work in advanced depletion methods. Edward Blandford (UNM) is performing measurements and modeling to characterize thermal-hydraulic performance of materials proposed for accident-tolerant fuels that will be evaluated in the first TREAT experiments. Lastly, Ben Forget at MIT is leading two research projects, both in development of advanced methods based on the Monte Carlo approach for improving TREAT modeling capabilities.

### Summary

**NCSU:** Research has focused on two tasks: (1) establish a neutronic model using the Serpent Monte Carlo code of the TREAT M2 calibration experiment according to the information given by Robinson (1985) and (2) establish a baseline graphite model that would enable calculation of the reference phonon spectrum for graphite. Task 1 has been completed, showing reasonable agreement between calculated and measured flux distributions in test fuel pins, but with a bias that is being investigated. The Task 2 model has been completed and tested using the density functional theory code VASP (i.e., Vienna Ab initio Simulation Package). The model shows good agreement with measurements of phonon dispersion relations. Completion of both of these tasks will support improved modeling and validation for TREAT.

**OSU:** During the first year, the collaborators demonstrated that Rattlesnake (i.e., the neutron transport solver in the Multi-Physics Object-Oriented Simulation Environment [MOOSE]) could be used to compute the phonon distribution in heterogeneous materials and that moments of this distribution could be used to predict thermal conductivity. Results for several test problems were generated and the Rattlesnake calculations were compared with values from literature. During the second year, the reactor analysis package MAMMOTH was used to complete steady-state calculations for the TREAT minimum critical core configuration. The fundamental neutronic properties were investigated, as well as the effects of spatial homogenization and angular discretization on power distribution, eigenvalue, and integral reaction rates. Preliminary diffusion constant treatments in highly anisotropic regions were developed. Current work involves investigation of self-optimizing, reduced-order systems of depletion equations. Collaboration between OSU and Professor Ben Forget at MIT has recently been

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<sup>2</sup> North Carolina State University

<sup>3</sup> Oregon State University

<sup>4</sup> University of New Mexico

established. This research has supported code validation and promises to provide new potentially powerful methods for depletion modeling.

**MIT:** Two areas of research at MIT also support TREAT modeling: (1) integration of the thermal neutron scattering of graphite in Monte Carlo simulations to facilitate sensitivity studies and uncertainty analyses, and (2) coupling of Monte Carlo methods with the MOOSE framework for coupled simulations. The methodology relies on two components: (a) functional expansion tallies for representing power distribution in continuous space, and (b) continuous material tracking for sampling neutron travel in a continuously changing medium. Under the first area, work has been completed by integrating pre-processing capabilities directly into the Monte Carlo simulations to facilitate generation of tables at all needed conditions. To improve versatility, MIT is pursuing the capability of sampling from thermal neutron scattering on-the-fly. For the second area, functional expansion tallies were implemented and tested using Legendre polynomials, whose coefficients are used to pass information to the finite element mesh of MOOSE, which greatly simplifies mapping. The first will improve understanding of uncertainties in TREAT graphite and the latter has already been integrated into INL work to allow coupling of the Serpent Monte Carlo code with MOOSE-based tools.

**UNM:** Wettability and surface roughness measurements for candidate accident-tolerant fuel (ATF) cladding samples have been performed using samples that were tested in water under different pressurized water reactor and boiling water reactor conditions. A pool boiling facility has been designed, fabricated, and assembled to perform experiments for collecting critical heat flux data on ATF cladding samples. A flow boiling loop has been constructed and tested for leakage and will be ready for critical heat flux testing after fabricating the heating elements. RELAP5-3D modeling and simulation of the water/Freon loop has been performed to determine experimental conditions. The measurements will support experiment design for early TREAT experiment with an ATF sample and will provide critical material and flow data needed for RELAP-7 simulation of those experiments.

## Benefits to DOE

All research projects described above are intended to advance the state-of-the-art for modeling and simulation of TREAT and its experiments. Much of the work will extend to a broader scope of reactor modeling. Therefore, this work very closely supports the DOE Nuclear Energy Modeling and Simulation Program and, specifically, the current TREAT modeling and simulation work package within the Nuclear Energy Modeling and Simulation Program. The various tasks, although diverse, are highly innovative and relevant to the DOE missions of transient testing and sustainability of the current commercial light water reactor fleet.

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Blandford, E. et al., 2016, “Development of an Electrical Capacitance Void Fraction Sensor for Accident Tolerant Fuel (ATF) Testing at the Transient Reactor Test (TREAT) Facility.”

Ellis, M., D. Gaston, B. Forget, and K. Smith, 2015, “Preliminary Coupling of the Monte Carlo code OpenMC and the Multiphysics Object Oriented Simulation Environment (MOOSE) for Analyzing Doppler Feedback in Monte Carlo Simulations,” *M&C 2015*, May 2015.

Ellis, M., D. Gaston, B. Forget, and K. Smith, 2015, “Preliminary Coupling of the Monte Carlo code OpenMC and the Multiphysics Object Oriented Simulation Environment (MOOSE) for Analyzing Doppler Feedback in Monte Carlo Simulations,” *M&C +SNA + MC 2015*.

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Ellis, M., D. Gaston, B. Forget, and K. Smith, 2016, “Preliminary Coupling of the Monte Carlo code OpenMC and the Multiphysics Object Oriented Simulation Environment (MOOSE) for Analyzing Doppler Feedback in Monte Carlo Simulations,” *Annals of Nuclear Energy*, invited

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### **Interns and Postdocs**

Interns: Jonathan Wormald, Nina Colby Sorrell, Anant Singhal, Anthony Alberti, Jackson Harter, Matthew Ryals, Carl Haugen, and Matt Ellis

Postdocs: Daniel LaBrier and Maolong Liu

### **Collaborations**

University: Massachusetts Institute of Technology, North Carolina State University, University of New Mexico, and Oregon State University

## 15-094—Evaluation and Demonstration of the Integration of Safeguards, Safety, and Security by Design

Jay Disser,<sup>1</sup> Janine Lambert,<sup>1</sup> Edward Blandford,<sup>2</sup> Edward Arthur,<sup>2</sup> Bobbi Merryman,<sup>2</sup> and Nicholas Osterhaus<sup>2</sup>

### General Project Description

The 3S by Design (3SBD) concept is designed to capture and potentially optimize inter-relationships between the safety, security, and international safeguards of a nuclear facility. This project is meant to provide nuclear reactor operators and designers with a cost-effective method of integrating safety, security, and safeguards considerations into reactor design and provide a risk-informed decision making platform for safety, security, and safeguards personnel. Because the Pebble Bed Fast High-Temperature Reactor (PB-FHR) design process is in its beginning stages, the system was chosen as a test case for a quantitative application of the 3SBD approach. Insights resulting from analysis are expected to be relevant to other advanced reactor designs, namely the gas-cooled pebble modular reactor and, potentially, low-pressure coolant reactor designs (e.g., the sodium-cooled fast reactor).

The 3SBD framework used for this research is notionally shown in Figure 1 (a complete 3SBD analysis of the PB-FHR would require an expanded framework that is beyond the scope of this project). The first step defines the pre-conceptual design of the facility of interest. The next step identifies off-normal scenarios, which cover potential initiating events and vulnerabilities across the 3S spectrum. During this project, RELAP5-3D is used to evaluate the response of the PB-FHR passive decay heat removal system while a simplified nuclear material accountancy model was developed to analyze the pebble fueling strategy.

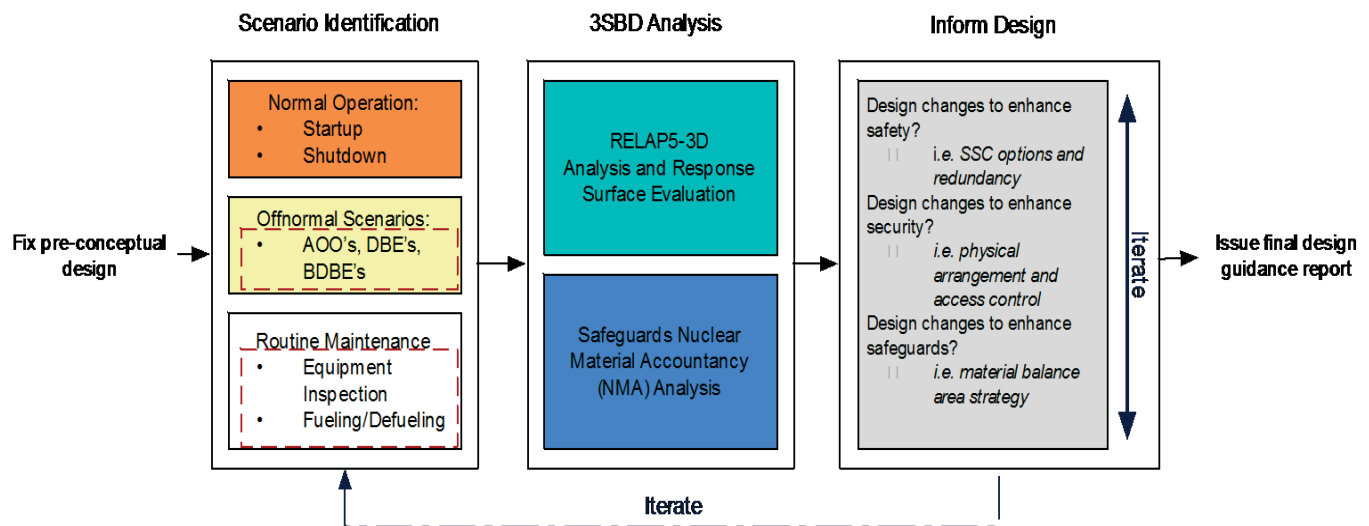


Figure 1. Notional schematic showing 3SBD approach.

### Summary

The University of New Mexico team modeled the PB-FHR in RELAP5-3D and analyzed the direct reactor auxiliary cooling system, which is a part of the reactor's decay heat removal system. The conclusions of the work showed that, in general, the response of the decay heat removal systems to partial blockages were fairly robust,

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requiring multiple high-percentage blockages to lead to large temperature excursions. In particular, blockages nearer the core (applied to the DHX primary coolant branch) had a much larger impact on early peak temperatures compared to blockages in the direct reactor auxiliary cooling system loop, although both scenarios lead to extended temperatures over time. This analysis was focused on reactor safety; however, security insights can be inferred to show which reactor systems are more sensitive to disruption from a physical protection or cybersecurity point of view.

In order to evaluate the safeguards significance of the PB-FHR, a nuclear material accountancy analysis was performed by using the projected material flows through the reactor system and the published uncertainties for existing instrumentation (Andreades et al. 2014). Three facility misuse scenarios were also examined. In conducting this analysis, an emphasis was placed on simplifying the system so the relative sensitivities of each area could be seen. Further sensitivity analysis and refinement of the model needs to be completed before a complete assessment of the nuclear material accountancy implications can be made. However, from this preliminary analysis, the most sensitive areas of the plant in regards to measurement uncertainty are fresh fuel storage and spent fuel storage, where the inspector and operator are going to be making measurements on large containers that contain thousands of pebbles in order to determine the mass of the nuclear material inside.

## Benefits to DOE

This project will place the United States in a stronger position when engaging other countries on 3SBD for critical nuclear infrastructure by providing a more objective method for 3SBD evaluation. This project will also establish the foundation to move 3SBD from a concept to practice. China is actively developing and demonstrating two pebble bed reactors. The Chinese PB-FHR is being developed by the Chinese Academy of Sciences and is expected to start construction in 2017. Although there has been a cooperative research and development agreement signed between the Chinese Academy of Science and Oak Ridge National Laboratory, very little effort in the research and development process has focused on safeguards and security considerations thus far. INL's leadership in thinking critically about using safety, security, and safeguards insights early in the design process can improve next-generation nuclear infrastructure.

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## **Interns and Postdocs**

Interns: Janine Lambert, Nicholas Osterhause, and Bobbi Merryman

## **Collaborations**

University: University of New Mexico

## 15-141—Interfacing MOOSE Components to Enhance Capability

Hongbin Zhang,<sup>1</sup> Carol Smidts,<sup>1</sup> Xiaodong Sun,<sup>1</sup> and Jinsuo Zhang<sup>1</sup>

### General Project Description

Fission products are released into the coolant system through fuel cladding defects or during severe accidents. This results in radioactive material being released into the coolant. The released fission products are deposited on the cladding walls and the coolant system and cause corrosion of fuel cladding and reactor structures. The coolant also reacts with the fuel through the defects and causes fuel oxidation. The purpose of this study is to develop a non-equilibrium chemical model for the assessment of chemical and corrosion reactions among coolant, cladding, and fuel. It also includes the assessment of hydrogen production and cladding oxidation. Within the context of the distributed test facility, project objectives are to investigate the connection between RAVEN/RELAP-7 and the distributed test facility (DTF) for instrumentation and control systems. A reduced-scale, hardware-in-the-loop (HIL) steam generator, water level control system and a full-scale nuclear power plant simulator will be used to interact with RAVEN. This will allow capabilities to be extended in the area of instrumentation and control validation and verification and HIL testing. In the area of thermal hydraulics, the objective of the task is to support two-phase flow modeling and validation activities by investigating the thermal non-equilibrium five-equation drift flux model (DFM) using the computational framework Multi-Physics Object-Oriented Simulation Environment (MOOSE). Work will be performed to improve the five-equation DFM formulation and improve/develop necessary closure relations.

### Summary

#### ***Release of Fission Products and Materials Corrosion***

The solubility of the fission product species, their kinetics properties, and the fission products release incorporated with the fuel oxidation model are under testing/development during the first year of the project. Because the thermodynamic data of some fission product species of the light water reactor are not easily available, rare earth fission products, including lanthanum, neodymium, and cerium, were first chosen to be studied. Their solubility in boric acid-lithium hydroxide solutions at room temperature and 40°C are studied. An electrochemical experiment has been set up for studying the Gibbs free energy, activity coefficient, and diffusion coefficient of the fission products in aqueous solution. A fuel oxidation model has been established using MOOSE/BISON, which takes into consideration hydrogen production, the defects' sizes, and their locations on the fuel cladding.

#### ***Thermal Hydraulics***

During this reporting period, development of the five-equation DFM was continued, specifically development of the constitutive equations. Compared to the four-equation DFM, a dispersed phase (i.e., gas phase) energy equation derived from the two-fluid model was added to comprise the five-equation DFM. The interfacial mass and heat transfer terms between the two phases are required to solve for the five-equation DFM in order for the five-equation DFM to be applied to thermal, non-equilibrium conditions in two-phase flow. Through a literature survey of available publications and system analysis code manuals, additional constitutive relations about the interfacial transfer models needed to close the five-equation DFM were investigated. In addition, the system of equations for the five-equation DFM is hyperbolic and well-posed. The five-equation DFM is being implemented into the MOOSE framework for a future benchmark.

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<sup>1</sup> Idaho National Laboratory

### **Distributed Test Facility**

The licensing process for RAVEN is nearing completion. An updated technology control plan has been approved for the terms of use and access to RAVEN. A secured network access to the INL repository is being set up to allow access to the source code. The compiled executable will be used in conjunction with the DTF on a local network to prevent unauthorized access to RAVEN. Fully using RAVEN's capabilities requires extension of the DTF framework to include failure modes for hardware components in the HIL system and the ability to trigger these failure modes. First, the failure modes of a valve in the turbine bypass system were considered. The failure-modes-and-effects-analysis method was used to analyze performance of the turbine bypass valve. Equivalent behavior rules for operational modes (i.e., plugging, internal leakage, internal rupture, external leakage/rupture, and spurious operation) are being used to study the effect of real failure modes and their influence on the system's performance. The equivalent failure modes are being implemented in the HIL setup. RAVEN will be able to set the status of a valve in the HIL system to one of these modes. This will be the first use of RAVEN within an HIL system. A full-scale nuclear power plant simulator has been considered for integration with RAVEN and/or DTF. First, the simulator source code was obtained and the appropriate variables that need to be replaced by values from the HIL system were determined. A tool has been developed to (1) search the desired system variables that need to be modified by external codes, (2) insert a subroutine into the source to overwrite that variable's internal value with an external value, and (3) recompile the simulator source code. The appropriate variables are then transmitted to and from the physical hardware to form a closed loop composed of the simulator and reduced-scale steam, general water level control system. This configuration also allows RAVEN to inject faults into the simulator, the HIL system, or both. Because the simulator models a full-scale system, the variables were appropriately scaled before being implemented by the reduced-scale hardware. Issues with a mismatch of scaling factors for input and output variables are being investigated. Further issues (e.g., handling slower-than-real-time simulations and restoring dynamic states of the HIL system after communication failure) are being studied.

### **Benefits to DOE**

This project benefits DOE by uncovering new thermodynamic data for fission products and extending the capabilities of MOOSE/BISON. This work will help obtain additional thermodynamic data for fission products that are released in the light water reactor coolant and study the mechanisms of fission product release.

Implementation of the MOOSE framework to solve the five-equation DFM will offer a practical and accurate tool to model transient two-phase flow phenomena in light water reactors. This work will help extend application of the MOOSE Framework in solving the non-thermal equilibrium two-phase flow model. The connection of RAVEN to the HIL setup at Ohio State University will be the first use of RAVEN in conjunction with physical components, thereby extending RAVEN's capabilities. The connection of RAVEN to the full-scale nuclear power plant simulator will similarly extend RAVEN's capabilities to interface with other complex software.

### **Publications**

#### **Presentations**

Aggarwal, R. and C. Smidts, 2016, "Interfacing MOOSE Components to Enhance Capability: Interfacing RAVEN with the Distributed Test Facility," Idaho National Laboratory – *National University Consortium Collaboration Projects Annual Review Meeting*, Idaho Falls, Idaho, August 16, 2016.

Shi, S. and X. Sun, 2016, "Interfacing MOOSE Components to Enhance Capability: Thermal Non-equilibrium Drift-Flux Model and its Implementation into RELAP7," *Idaho National Laboratory – National University Consortium Collaboration Projects Annual Review Meeting*, Idaho Falls, Idaho, August 16, 2016.

Xue, S. and J. Zhang, 2016, “Interfacing MOOSE Components to Enhance Capability,” Idaho National Laboratory – *National University Consortium Collaboration Projects Annual Review Meeting*, Idaho Falls, Idaho, August 16, 2016.

### **Interns and Postdocs**

Interns: Mike Pietrykowski, Rachit Aggarwal, Huijuan Li, Bobak Rashidnia, and Sha Xue

Postdoc: Shanbin Shi

### **Collaborations**

University: Ohio State University

## 15-142—New In-Core Neutron Diagnostics

Sebastien Teyssyre,<sup>1</sup> Adam Hecht,<sup>2</sup> and Jean-Claude Diels<sup>2</sup>

### General Project Description

This work aims to use optical methods for measuring neutron dose to optical crystals. Neutron damage causes changes in material structure following atom recoils and displacements. These changes in structure lead to changes in the index of material refraction, which we can measure. The goal is to demonstrate that we can use this optical method for live-time monitoring of reactor neutron flux.

### Summary

After initial funding was received (June 2015), equipment (including the CaF<sub>2</sub> crystals to be used for this work) was procured and installed and work was initiated. Two students are involved in this project.

This year, the activity focused on characterizing the repeatability of measurements with known crystals and on generating crystals with increasing irradiation damage. Work also has been done on the effect of gamma irradiation on optical properties. Finally, efforts in modelling the evolution of a crystal under irradiation were undertaken.

### **Repeatability Measurements**

We needed to confirm there were no repeatability issues when running measurements, especially since we move crystals (e.g., for additional irradiation). Comparison between single crystals showed high repeatability, with point-by-point comparisons for different angles and for different cavity lengths giving a high-frequency/low-frequency ratio of about 62 (i.e., a function of the resonance order in the cavities) and a standard deviation of the mean of  $10^{-7}$  (i.e., **eight orders of magnitude of precision in repeatability**).

In conclusion, Because of the high repeatability, we can perform continuous monitoring and measure a change in the refraction index or we can take the crystal out and perform discretized irradiations. This fact not only supports the feasibility of the study in the laboratory but is also valuable for future practical applications.

### **Irradiations with a Plutonium-Beryllium Neutron Howitzer Source**

We performed several irradiations (irradiation flux measured  $5 \times 10^4$  n/cm<sup>2</sup>s at the sample location) on a sample for a total irradiation time of 153 hours, with testing between individual irradiations. Initial results suggest an evolution of the refraction index (i.e., qualitative measurements). Data analysis is in progress.

### **Examination of Electron Effects – Electron Displacements and Color Center Formation**

CaF<sub>2</sub> is very well calibrated for optically stimulated luminescence and has an American Society of Testing Materials standard (ASTM E2450-11) for use with gamma irradiation and mixed gamma/neutron fields. Because we are never in a neutron environment that is gamma free, we can use this as a second approach to understanding neutron fields. We are examining optical transmission spectra through crystal samples, starting with comparisons between samples for higher-precision measurements. Unirradiated samples have been tested and irradiated samples are starting to be used.

### **Simulations Using Molecular Dynamics**

Because the evolution of microstructure leads to evolution of the observed optical properties, there is a need to determine evolution of microstructure under irradiation to better understand this relation and to be able to predict

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<sup>2</sup> University of New Mexico



the crystal response as fluence increases. The student undertook evolution modelling of the microstructure of a CaF<sub>2</sub> crystal under irradiation using Large-scale Atomic/Molecular Massively Parallel Simulator simulations.

## Benefits to DOE

This new capability for neutron dosimetry will be relevant to use for a number of applications, including monitoring of irradiation experiments, conditions within multiple nuclear reactor types, separations technologies, and model validation. Through this capability, strides can be made in the areas of reactor reliability and safety and lifetime extension through the new diagnostic capabilities proposed here.

The novel detection capability will assist in a more comprehensive understanding of the radiation damage occurring within the reactor at specific locations and times. This technology can be applied in a broader sense to other systems, including model validation. Overall, this will enable a better understanding of radiation damage and microstructural evolution in fuels and materials because real-time diagnostics for neutron damage will be developed.

## Publications

Masuda, K., E. I. Vaughan, L. Arissian, J. P. Hendrie, J. Cole, J.-C. Diels, and A. A. Hecht, 2015, “Novel Techniques for High Precision Refractive Index Measurements, and Application to Assessing Neutron Damage and Dose in Crystals,” *Nuclear Instruments and Methods A*, Vol. 784, pp. 198.

## Presentations

Hendrie, J., L. Arissian, K. Masuda, A. Hecht, and J.-C. Diels, 2015, “Nested Fabry-Perot in Mode-Locked Lasers to Monitor Minute Changes of Index,” *CLEO: Science and Innovation*, San Jose, California, May 10 through 15, 2015, ISBN: 978-1-55752-968-8.

## Interns and Postdocs

Interns: Sara Pelka and Joe Morris

## Collaborations

University: University of New Mexico

## 15-144—Investigation of Sonication-Assisted Electrolytic Reduction of Used Oxide Fuel in Molten Salt

Shelly Li,<sup>1</sup> Robert O'Brien,<sup>1</sup> and Haiyan Zhao<sup>2</sup>

### General Description

Electrolytic reduction is an integral step in pyroprocessing to treat used fuel from light water reactors and other metal oxides. During electrolytic metal oxide reduction, oxygen ion diffusion from fuel particulates inside the cathode basket to the anode is the limiting step, which leads to low current efficiency, long operating hours, and re-oxidation of reduced metal fuels. Extensive research and development activities have been conducted in the past decade to improve the oxygen diffusion process associated with electrolytic reduction of used light water reactor fuel in high-temperature molten salts. Sonication is a mature technology used to improve a variety of chemical/physical processes, including chemical reactions and mass transports. However, sonication technology has not been explored to improve the electrolytic reduction process. This project provides a great opportunity to investigate the feasibility of improving electrolytic reduction of used oxide fuel in molten salts using ultrasound techniques.

### Summary

In this project, a sonication apparatus will be applied to an electrochemical cell with molten LiCl-Li<sub>2</sub>O at 650°C. The effect of sonication for improving the performance of electrolytic reduction of surrogate metal oxide (such as TiO<sub>2</sub> or NiO) will be studied at the Center for Advanced Energy Studies (CAES) at the University of Idaho. Then the sonication-assisted electrolytic reduction of depleted UO<sub>2</sub> will be investigated at both the University of Idaho/CAES and the Fuel and Applied Science Building at the Materials and Fuels Complex of INL.

During FY 2016, the equipment for electrolytic metal oxide reduction has been setup within an argon-purged glove box at CAES. TiO<sub>2</sub> was selected to be a UO<sub>2</sub> surrogate for simplicity and cost effectiveness. The initial experiments have focused on establishing the baseline for the current efficiency of TiO<sub>2</sub> reduction without sonication. The TiO<sub>2</sub> electrolytic reduction at 650°C in LiCl-Li<sub>2</sub>O was investigated. The cyclic voltammetry technique was applied to determine the stable operational window for TiO<sub>2</sub> reduction. The constant potential with intervals was applied for TiO<sub>2</sub> reduction. A weight-loss method and post-reduction analysis, including x-ray diffraction and transmission electron microscopy, were developed to quantify the reduction extent and identify the phases in processed TiO<sub>2</sub> samples. It has been found that it was very challenging to reduce TiO<sub>2</sub> with a detectable quantity within a reasonable time (i.e., about 5 hours for 2 g of TiO<sub>2</sub>). Reduction products mostly consisted of the lithium titanates LiTiO<sub>2</sub>, Li<sub>2</sub>TiO<sub>3</sub>, and LiTi<sub>2</sub>O<sub>4</sub>. This study revealed a detailed electrolytic reduction mechanism for TiO<sub>2</sub>. Future work will focus on using other surrogates such as NiO to establish the baseline of current efficiency prior to employing sonication-assisted electrolytic reduction. An experimental mockup was constructed to demonstrate that the sonicator probe can fit into the crucible and operated alongside the electrode assembly without modifying the setup, which ensures the result is directly comparable without and with sonication. Working control document for performing electrolytic reduction with depleted UO<sub>2</sub> is being reviewed. A detailed experimental plan, procedures, and safety measures are under preparation.

### Benefits to DOE

Agitation through sonication is expected to accelerate oxygen ion diffusion, improve the contact between fuel particulates with the cathode, and drive trapped O<sub>2</sub> molecules diffusing out of the packed fuel bed into the

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<sup>1</sup> Idaho National Laboratory

<sup>2</sup> University of Idaho

adhesive molten salt. Should the proposed research proceed as expected, it will successfully resolve one of the key needs associated with scaled-up pyroprocessing. The proposed research will expand expertise in the electrolytic reduction of used light water reactor fuels for pyroprocessing and further establish the INL as a world leader in innovative fuel cycle technology. The proposed research is also directly applicable to the projects for recycling rare earth elements under the Critical Material Institute. Should this proposal yield technical advances, funding would be sought through the DOE Fuel Cycle Research and Development Program, as well as the Critical Material Institute and DOE Nuclear Energy University Program. The research team will seek to protect intellectual property and publish information in the interest of the laboratory.

## **Publication**

Song, E., 2016, "Challenges with Electrochemical Metal Oxide Reduction in Molten Salts," Capstone Project, nuclear engineering masters student, August 2016.

## **Presentation**

"Effect of Electrolyte Purity on  $\text{TiO}_2$  Electrolysis in Molten  $\text{LiCl-Li}_2\text{O}$ ," student poster, *ANS Winter Meeting*, Las Vegas, Nevada, 2016.

## **Interns and Postdocs**

Interns: Eric Song, Jeremiah Dustin, Jieun Lee, and Seungje Oh

## **Collaborations**

University: University of Idaho

## 15-145—Advanced Neutron and X-Ray Imaging at TREAT

James K. Jewell<sup>1</sup> and Tony Hill<sup>2</sup>

### General Project Description

This project is focused on development of advanced imaging options for the future Transient Reactor Test (TREAT) experimental program with an aim of developing a suite of possible imaging capabilities that will extend the scientific and engineering reach of TREAT to include a lower-length scale scientific testing program.

### Summary

This project plan starts with a broad analysis to identify spatial and temporal requirements for achieving the overarching goals of the program and the specific requirements as determined by potential users. A workshop was held September 2nd and 3rd in the Center for Advanced Energy Studies auditorium that brought local experimenters and modelers together in order to begin identifying high-priority validation needs. The discussion focused on development of appropriate sensitivity studies under the Multi-Physics Object-Oriented Simulation Environment (MOOSE) framework that can support future cost-benefit analyses. The discussions were not limited to central position experiments, but included ideas for evolving the TREAT core to include a lower-length scale irradiation position near the periphery that can be used parasitically, particularly during the time between central position irradiations, which could extend and complement the traditional TREAT experimental program. Discussions also focused on development of closer collaboration between modelers and experimentalists to properly design and analyze experiments of high value, because no organizational system is in place to cultivate and manage this relationship. Sensitivity studies are needed that provide a quantitative basis (i.e., benefit) for experiments (i.e., cost) and requires initial uncertainty evaluations in existing models and covariance development. Experimentalists need a high-fidelity simulation tool for the experimental regions that support instrument/experimental design. The need for experimental analysis and data mining in MOOSE was discussed for use of large-field scanning electron-microscope-based “Big Data” development.

Spatial energy deposition measurements can be considered critical for advancing the scientific impact of nearly all TREAT experiments. A rather simple system has been studied that would provide (a) the previous burn-up profile along the axis of the fuel rod as it is being inserted into the TREAT reactor, and (b) the energy deposited by TREAT along the axis of the rod as it is being retrieved from the reactor, thus determining the power coupling of the rod as a function of its length. This can be accomplished with the addition of an experimental collar or extension to the fuel cask that is used to transport the fuel to and from the reactor. The extension would be positioned between the bottom of the cask and the top of the reactor. The extension will provide the shielded experimental access needed to “view” the fuel as it is moved into and out of the reactor. A simple horizontal slit (or slits) in the collar that runs from the inner diameter to the outer diameter will allow gamma rays to escape the central position and be measured by a hyper-purity germanium system located on the outer diameter. The fuel can be stepped down from the cask, through the collar, and into the reactor, allowing gamma spectra to be measured as a function of length along the fuel rod. Long-lived isotope ratios will be used to determine previous history; short-lived isotopes will be used for measuring the TREAT coupling. These measurements could significantly reduce the uncertainties associated with the specific irradiation conditions as a function of position within the test vehicle. The fluctuations in the neutron flux cannot be specifically predicted for each experiment; therefore, measurements must be carried out that quantify these variations in order to maximize the scientific impact. Signal-to-noise calculations have been estimated based on drawings of the multi-SERTA test vehicle, which not only shielded and scattered gamma, but increased backgrounds due to activation. However, the neutron

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<sup>1</sup> Idaho National Laboratory

<sup>2</sup> Idaho State University

flux shape for the TREAT experiment position has only recently been estimated (poorly) and has not been incorporated into this study yet. GEANT4-based simulations are under development to identify the benefits of specifically designed hyper-purity germanium “clusters” that are spectrally more efficient with lower background levels in the Compton continuum. This simulation will also provide first estimates of the intrinsic backgrounds from Compton scattering within the dense nuclear fuel, which represents an irreducible background in the system and directly impacts the signal-to-noise requirements for such a system.

An advanced, single gamma emission, computed tomography system can provide true three-dimensional isotopic distributions in fuel rods or pellets for determining axial burnup and power profile, percent fission gas release, and isotopics of fission gases, as well as isotopic distributions within the fuel. A dedicated system has been envisioned that could be located above the hot cell in the loop insertion cell. There have been a number of technological advances in computing, electronics, and gamma detector design since the era of computed tomography began. A new simulation tool was developed specifically for carrying out basic sensitivity studies on the tomographic reconstruction code and eventually on global design parameters. Existing three-dimensional codes (such as MCNP and GEANT) are expensive calculations given the overall inefficiency for the gamma detection system, which is reduced to a highly restricted, two-dimensional problem. The new two-dimensional simulation code performs analytic line integrals across the idealized object in the measurement direction with an adjustable step granularity in the perpendicular direction that are summed to represent the flux of gammas through an adjustable collimation system. The new simulation code was used to quickly and efficiently generate data that were processed by modern computed tomography codes. It turns out that the intrinsic two-dimensional resolution is fixed by the gamma collimation slit width, which will be about 1 mm in a realistic system. The potential for high resolution (i.e., about 20 micron) remains a possibility with 1-mm apertures, but will require significant effort in developing a new Bayesian unfolding algorithm (for instance) that maximizes the presented data in a true three-dimensional space that accounts for variations in fuel density, diameter, and curvature of rods. This approach would certainly require modern computing infrastructure and approximately 4 full-time equivalent years to develop the code. A more efficient unfolding algorithm will be needed to carry out further design sensitivity studies.

Approaches for validating the thermodynamic behavior of TREAT fuel are also being investigated. Under the assumption that fuel remnants are available, we foresee a set of experiments using highly instrumented fuel shapes that will be used as targets to provide critical feedback to the MOOSE-based verification and validation effort. One candidate is the central irradiation of small, bare TREAT fuel specimens that are highly instrumented for temperature mapping over the entire surface. The primary instrumentation challenges of this experiment are time response of thermocouples, which must have a time constant much shorter than a fast transient time scale (100 ms), and the coupling technique for a large number of measurement points, which could be a hundred or more. Research has demonstrated that thin film thermocouples (i.e., 125-um layers) are capable of response times below a microsecond, making them prime candidates for fast pulse studies. These thermocouple layers are usually deposited by evaporation or sputtering techniques directly onto the surface of a sample using photolithography masks. The two layers are offset so each pad has primary contact with the surface, along with a small overlapping area that forms the Seebeck junction. Each pad is also used for contacting the system with wires for voltage measurements by a remote data acquisition system. Complications arise in this application due to the radioactive nature of the specimen and the conductivity of the fuel surface. We are studying different approaches for simplifying the attachment of thin-film thermocouples and ways for electrically isolating junction pairs from the fuel specimen surface or including graphite in the thermocouple circuit.

## Benefits to DOE

This work supports the DOE Office of Nuclear Energy Roadmap directive for science-based, independent effects measurements. The creation of advanced imaging and monitoring capabilities for fuel materials under transient

power irradiations with line of sight access would be the first of its kind. It represents a novel approach to the detailed study of fuel at multiple length scales. This approach fulfills the vision proposed in the Office of Nuclear Energy Roadmap for the future of a science-based approach to fuels development research; it impacts all four main research and development objectives.

### **Publications**

Hill, T., 2016, “Advanced Neutron and X-Ray Imaging at TREAT,” *LDRD Mid-Year Review*, June 28, 2016.

### **Collaborations**

University: Idaho State University



## 16-003—Recycling of Tantalum-Containing Waste Materials to Recover Tantalum Metal

Prabhat K. Tripathy,<sup>1</sup> Michael R. Shaltry,<sup>1</sup> and Jerome Downey<sup>2</sup>

### General Project Description

The specific objective of this project is to develop a high-temperature electrochemical process with a view to reclaiming tantalum metal from its primary and secondary resources. While the primary source is its oxide ( $\text{Ta}_2\text{O}_5$ ), the secondary resources constitute manufacturing waste and spent/discarded material from industry/user sources. The focus for the first year was to carry out fundamental measurements pertaining to oxidation behavior of pure tantalum metal in a variety of conditions that are usually encountered in industrial applications and the electrochemical behavior of the oxidized metal in a molten salt. The oxidation behavior of the metal was studied both in static air and under the flowing gas atmosphere in a thermobalance. Electrochemical measurements were carried out in both (anhydrous and high purity)  $\text{LiCl}+1 \text{ wt. \% Li}_2\text{O}$  and  $\text{CaCl}_2+1 \text{ wt. \% CaO}$  melts. Chemical analysis of the reduced metal showed that the electrochemical process could transform the tantalum pentoxide ( $\text{Ta}_2\text{O}_5$ ) to high-purity tantalum powder in just one unit operation.

### Summary

Although tantalum is not regarded as a precious metal, its intrinsic value has been the driving force to warrant its recycling, which is why the world-wide tantalum industry is trying to develop a closed-loop concept for production of tantalum. The United States has no tantalum mining industry and, as a result, imports all of its tantalum source materials. Primary and secondary sources of tantalum include tantalite and columbite ores that are being mined mostly in Australia, Brazil, Canada, and the Democratic Republic of the Congo and from tantalum-bearing tin slags (both low and high grades), respectively. The extraction of tantalum from its ore bodies involves two broad unit operations: dissolution of the ore in hydrofluoric acid followed by solvent extraction with methyl isobutyl ketone (also known as MIBK) extractant. This procedure efficiently recovers tantalum in a form that then can be further processed into tantalum oxide and potassium fluotantalate. The latter is then subsequently reduced with sodium to produce tantalum powder, which acts as the feedstock for production of tantalum wire, sheet, alloys, and capacitor-grade metal powder. The focus of this project is to develop an electrometallurgical process for production of tantalum metal from its oxide, as well as develop a one-step purification process for refining the oxidized tantalum scrap/waste materials without converting them to tantalum pentoxide. The third objective is to apply the electrometallurgical process to prepare and purify a variety of other engineering metals/alloys.

Figure 1 shows the scanning electron microscope photograph of a discarded tantalum capacitor that contained 5 to 46 wt.% oxygen. Spectrum 6 and 7 contained tantalum as high as 50% and 76%, respectively. These types of oxidized materials can very well be refined by a suitable electrometallurgical treatment. Figure 2 shows the oxidation pattern of pure tantalum metal in three different environments (i.e., flowing air, oxygen, and argon plus hydrogen mixture).

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<sup>1</sup> Idaho National Laboratory

<sup>2</sup> Montana Tech of the University of Montana

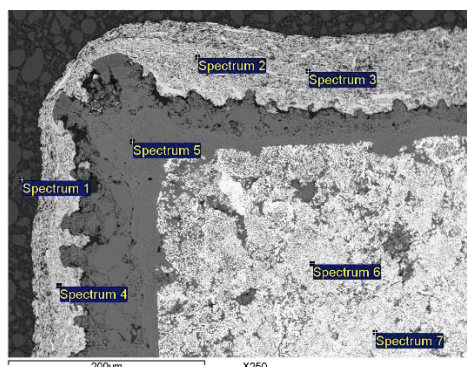


Figure 1. Scanning electron microscope image of a waste tantalum capacitor.

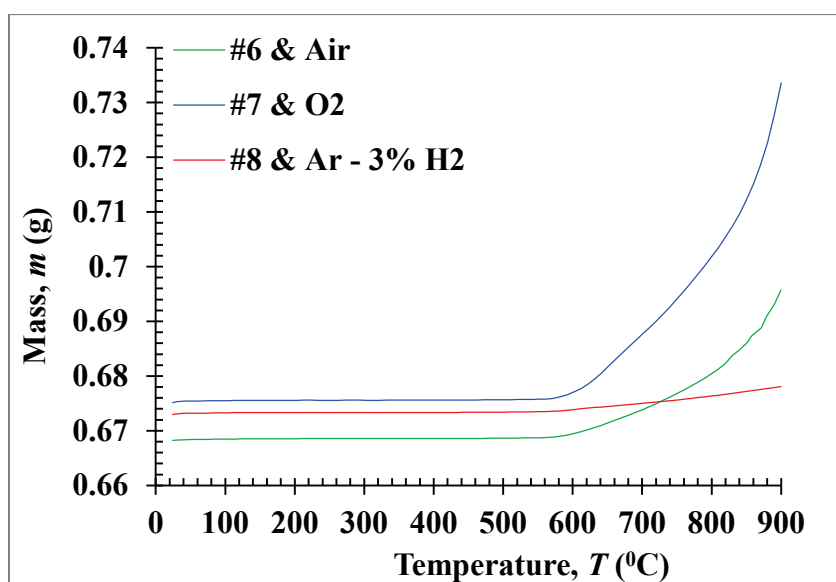


Figure 2. Mass gain profiles in three different atmospheres.

As expected, it can be clearly seen from Figure 2 that tantalum undergoes much faster oxidation (to  $Ta_2O_5$ ) in pure oxygen, than in static air, at a temperature higher than about 600°C. Similar to the case of molybdenum, oxidation kinetics of tantalum (to its oxide) is extremely sluggish in a slightly reducing atmosphere, even when heated to about 900°C. Another interesting observation was the occurrence of relatively lower oxidation rate at temperatures less than 600°C, even in static air. In fact, when heated in air up to a temperature of about 300°C, tantalum gets barely oxidized. These experiments indicate that significant oxidation of pure tantalum can be prevented if the metal is used at a temperature less than 300°C. Even better control on oxidation behavior can be accomplished under a slightly reducing atmosphere.

Subsequent electrochemical measurements performed on the oxidized wire indicated that upon polarization in a pool of molten lithium chloride electrolyte, tantalum got reduced to tantalum metal in just one step (Figure 3). The first redox couple (between approximately 0.6 to -0.45 V) was indicative of the oxidation and reduction of  $Ta^{+5}$  to  $Ta^0$  (Figure 3). The second couple (between approximately -1.6 to 1.2 V) was due to lithium deposition and stripping.

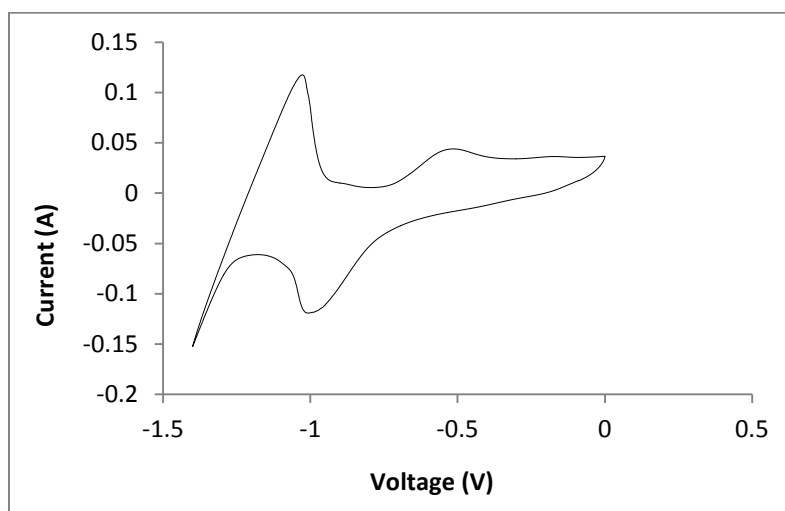


Figure 3. Cyclic voltammetric measurement in the  $\text{LiCl-Li}_2\text{O}$  electrolyte: scan rate =  $100 \text{ mVs}^{-1}$ , operating temperature =  $650^\circ\text{C}$ ; working electrode: oxidized tantalum wire (1-mm diameter); counter electrode: molybdenum coil; reference electrode: glassy carbon rod (3-mm diameter and 100-mm long); and working area surface area =  $0.5 \text{ cm}^2$ .

Electrochemical reduction of the sintered oxide pellet was performed in a calcium chloride melt at  $900^\circ\text{C}$  by cathodically polarizing the oxide pellet (Figure 4) against an oxygen-evolving electrode for durations up to 31 hours. The applied cathode potential during the reduction experiment was in the range of -1.8 to -1.9 V versus glassy carbon (pseudo reference) electrode. The current versus time profile showed an initial rise in the current followed by its smooth decline as the reduction reaction progressed with time. The initial peak and residual current values were 0.29A, 0.54A, and 0.14A, respectively. The cell potential was in the range 2.6 to 2.9 V. Measurement of the residual oxygen content in the reduced pellet (Figure 5) suggested that it could be possible to reduce the oxygen content from an initial value of 18.1% (in  $\text{Ta}_2\text{O}_5$ ) to 0.12% (in the reduced pellet), which is a 99.3% reduction.



Figure 4. Air-sintered oxide precursor prior to its electrochemical reduction (oxide powder size =  $<325$  mesh, sintering temperature =  $975^\circ\text{C}$ , and duration = 5 hours).



*Figure 5. State of the reduced pellet, partial silvery white color on the surface of the reduced pellet was solidified calcium chloride (electrolysis temperature = 900°C, electrolysis duration = 31 hours).*

## Benefits to DOE

One of the missions of INL is to develop knowledge, research, and manufacturing facilities in order to be able to advance knowledge in the field of efficient manufacturing to secure U.S. competitiveness. Another mission is to develop effective recycling strategies for critical and strategic metals. If successful, the proposed project will provide opportunities to attract national and international collaborations both from academia and industries. Thus, it is believed that the proposed scope of the project will directly contribute to furthering INL's goals and mission.

## Publications

Chorney, M. P., B. P. Hurley, P. K. Tripathy, and J. P. Downey, 2017, "Effects of Oxide Precursor Preparation Parameters on the Electrochemical Reduction of Tantalum Pentoxide ( $\text{Ta}_2\text{O}_5$ ) in Calcium Chloride Melt," *146<sup>th</sup> Annual Meeting & Exhibition, The Mineral, Metals and Materials Society (TMS)*, San Diego, California, February 26 through March 2, 2017.

Tripathy, P. K., G. L. Fredrickson, and T. A. Todd, 2016, "Use of a Monolithic Noble Metal as an Oxygen Evolving Electrode in Molten Salts," *eIDR* to be submitted.

Tripathy, P. K., M. R. Shaltry, and G. L. Fredrickson, 2016, "Oxidation Behavior, Microstructural Features and Phase Analysis of the Oxidized Tantalum Metal," to be communicated.

## Interns and Postdocs

Intern: Maureen Chorney

## Collaborations

University: Montana Tech of the University of Montana

## 16-009—Change Detection System for Nuclear Applications

Troy Unruh,<sup>1</sup> Greg Lancaster,<sup>1</sup> Michael Overton,<sup>1</sup> Walter Williams,<sup>1</sup> Jeff Brower,<sup>1</sup> and Ken Thomas<sup>1</sup>

### General Project Description

The focus of this effort is to conceptualize and design a set of systems for performing image analysis that will aid researchers and workers at nuclear facilities in identification, analysis, and tracking of materials and anomalies. This project developed an advanced image alignment capability called Change Detection Systems (CDS). This innovative software has been widely used in national security-related applications and is globally considered to be the preeminent system for image change detection. CDS has been deployed to numerous government agencies, in addition to receiving two patents and winning two Research and Development 100 Awards. When deployed for nuclear applications, CDS will transform the way work is accomplished by leveraging powerful computer vision techniques for the identification and analysis of objects/areas not currently available to the nuclear reactor community.

### Summary

Performed initial scoping research prior to developing a nuclear-focused version of CDS for integrating into daily operations at nuclear facilities. Our team interfaced with the appropriate researchers, program leads, and workers in Nuclear Science and Technology and Nuclear Operations at the ATR Complex, the Materials and Fuels Complex, and commercial nuclear power plants to further nuclear-focused development activities of CDS. Some examples of the applications that we evaluated include the Reduced Enrichment Research and Test Reactor 12 plates examined for blister formation during annealing; Neutron Radiography Reactor radiographs; Kijang Research Reactor fuel element condition; visual and thermal electronics assembly analysis; and a nuclear facility control panel from Palo Verde. Other areas we identified that would benefit with the inclusion of CDS image analysis techniques include facility work flow operations, independent remote verifications, work zone inspections in contamination areas, normal versus abnormal status indications, damage analysis (e.g., water hammer and seismic), thermography of live electrical connections, and tracking of fresh boron stains. Our research has shown that CDS will transform the way work is accomplished by leveraging powerful computer vision techniques for identification and analysis of objects/areas not currently available to the nuclear community. This has led to interest from potential commercialization partners and follow-on funding from DOE. The DOE Technology Commercialization Fund (see Figure 1) is being used to develop a nuclear-focused version of CDS called Apollo that will include thermal imaging capabilities on a smartphone; the Lab-Corp program is being used to better understand the commercial potential for such a product.

### Benefits to DOE

This project has led to the following two follow-on research and commercialization opportunities:

- \$125K — DOE Technology Commercialization Fund, “Commercialization Research and Development of Change Detection Systems for Nuclear Applications” (Figure 1)
- \$75K — Lab-Corps Team, “Change Detection Systems for Nuclear Applications.”

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<sup>1</sup> Idaho National Laboratory



Figure 1. Recently awarded DOE Technology Commercialization Fund Proposal.

## Publications

### ***Presentations***

Unruh, T., K. Thomas, M. Overton, and G. Lancaster, 2015, “Change Detection System (CDS) for Nuclear Applications,” *LWRS II&C Utility Working Group EPRI Strategy Group on Productivity Improvements through Advanced Technology*, Idaho National Laboratory, August 11 through 13, 2015.

### **Invention Disclosures, Patents, Copyrights**

Patent 7643703, “Image change detection systems, methods, and articles of manufacture.”

Patent 8351674, “Image portion identification methods, image parsing methods, image parsing systems, and articles of manufacture.”



## 16-010—Development of a Fully Coupled Radiation Damage Production and Evolution Simulation Capability

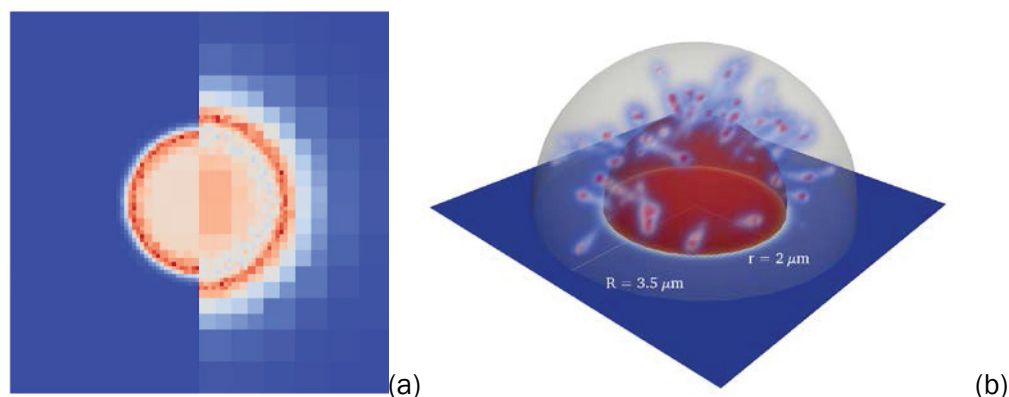
Daniel Schwen,<sup>1</sup> Sebastian Schunert,<sup>1</sup> Javier Ortensi, Xianming Bai,<sup>2</sup> and Yongfeng Zhang<sup>1</sup>

### General Project Description

Nuclear reactor components experience radiation damage due to energetic particle radiation. This creates defects in regular atomic lattices (such as vacancies and interstitials), causes disorder in ordered compounds, and redistributes material, impacting the microstructure evolution, which ultimately determines the macroscopic material properties. In this project, we aim to couple the simulation of radiation damage (MyTRIM), microstructure evolution (MARMOT), nuclide transmutation, and neutronics (MAMMOTH). The final deliverable will be a spatially resolved damage calculation capability with ballistic mixing informed by MyTRIM, transmutation in MARMOT informed by MAMMOTH, and residual defect densities implemented using the phase-field formalism.

### Summary

We completed coupling of the microstructure code MARMOT and MyTRIM in a flexible and highly efficient manner using a rasterization approach yielding maximum parallel scalability. MARMOT's FEM mesh is overlaid on the MyTRIM geometry. Material properties are computed on each FEM element and broadcast to each compute node for the MyTRIM simulation because collisions can span the whole domain. MyTRIM cascades parallelize over multiple MPI processes and threads.



*Figure 1: a) Xenon resolution from a gas bubble into the  $\text{UO}_2$  matrix (xenon depletion on the left and xenon enrichment on the right) around a 2-nm xenon bubble. b) Fission fragment tracks in graphite around a  $\text{UO}_2$  fuel particle with a radius of  $2\ \mu\text{m}$ . Tracks extend into a spherical volume with a radius of  $3.5\ \mu\text{m}$ .*

We demonstrated that the rasterization approach for coupling microstructure evolution and binary collision Monte-Carlo is feasible based on a  $\text{UO}_2$  xenon bubble benchmark showcasing the phenomenon of gas resolution, which is an important parameter for fission gas release models. The simulation comprises 500 cascades initialized by 200-keV xenon recoils (Figure 1a).

The neutron flux field is the driving force of radiation damage; therefore, it is vital for an accurate simulation of radiation-damage cascades. Within this project, we implemented the capability of computing the (angular) nuclide

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<sup>2</sup> Virginia Tech

recoil and nuclide fission rates that are used to construct PKA probability density functions. For sampling PKAs from fission reactions, ENDF data have been imported, incorporating the distribution of mass and the charge number of the fission products. ENDF data accurately represent the fission product distributions of more than 20 different fissionable targets for neutrons ranging from thermal energies (less than 1 eV) up to fast neutrons (20 MeV).

The utility of the coupled neutronics-binary collision Monte-Carlo simulation is demonstrated on a Transient Reactor Test Facility (TREAT) fuel particle. A TREAT fuel particle's size is of the same order as the range of fission fragments; therefore, the fission energy deposition around it is neither uniform nor confined to the fuel (i.e., a fraction of the energy is deposited in the graphite matrix). We will use MyTRIM to inform a heat conduction problem's source term during a TREAT transient calculation. Graphite vacancies developing around a spherical fuel particle with a radius of 20  $\mu\text{m}$  are depicted in Figure 1b. This will allow for a more detailed understanding of the mechanisms of thermal feedback translating into a more accurate prediction of peak power, position, and total deposited energy during a TREAT transient.

### Benefits to DOE

Understanding radiation damage is essential to advancing fuel performance. However, to understand radiation damage fuel performance must be viewed in a coupled, multi-physics setting. The benefit of this project is development of the first tool for comprehensive studies of radiation damage and its evolution in a multi-physics environment. It will advance our understanding of the microscopic mechanism governing the macroscopic behavior of fuels and structural materials, which will impact the ATF and Light Water Reactor Sustainability Programs. This project has already found application for the TREAT reactor and directly impacts INL's mission.

### Publications

#### ***Presentations***

Schwen, D. and S. Schunert, 2016, "Coupling Radiation Damage from Binary Collision Monte Carlo to Phase Field Microstructure Evolution," conference presentation INL/CON-15-35718, TMS 2016, Nashville, Tennessee, February 16, 2016.

Schwen, D. and S. Schunert, 2016, "Coupling Radiation Damage from Binary Collision Monte Carlo to Phase Field Microstructure Evolution," accepted abstract, MMM 2016, Dijon, France, October 12, 2016.

### Interns and Postdocs

Intern: Pedram Ghassemi

### Collaborations

University: Virginia Tech

National Laboratories: Los Alamos National Laboratory

## 16-013—Micromechanistic Approach and Critical Experiments for Quantitative Predictions of Delayed Hydride Cracking in Zirconium Alloys

S. B. Biner,<sup>1</sup> P. Chakraborty,<sup>1</sup> D. Schwen,<sup>1</sup> and S. Teyssyre<sup>1</sup>

### General Project Description

This project aims to develop a micromechanistic phase-field model, validated with critical experiments, for quantitative predictions of delayed hydride cracking in zirconium alloys.

### Summary

Progress on this project is summarized in the following two sections.

#### **Modeling and Simulation Studies**

A phase-field model for  $\delta$ -hydride evolution in zirconium alloys has been developed. The model correctly incorporates the misfit strains between the hydrides and the matrix phases on the evolution of the interfacial energy, leading to formation of the needle shape hydride phases for three different hydride invariants (see Figure 1).

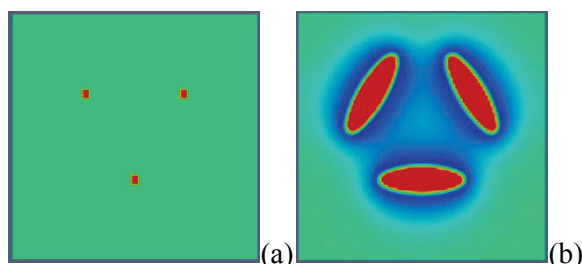


Figure 1. Evolution of three different hydride invariants from circular seeds. The hydrogen concentration is 0.1 in the matrix and 0.5 in the precipitate, with (a) the initial seed and (b) the final configuration of the precipitates.

In addition, incorporation of the thermodynamic database, based on the phase-equilibrium diagram, is currently being pursued for quantitative analysis. This approach enables us to model dissolution/precipitation under thermal cycling (see Figure 2).

A FEM code is under development to study the hydrogen flux to the crack tip under mix-mode loading conditions. The main objective of this effort is to elucidate two competing and conflicting theories/models that have been developed for delayed hydride cracking.

#### **Experimental Studies**

We have acquired pure zirconium in bar form to carry out the planned delayed hydride cracking experiments. We also acquired some Zircalloy-2 alloy in tube form and Zircalloy-4 alloy in plate form. In addition to the electrolysis route, we are also establishing procedures for introducing hydrogen into the crack growth specimens in a furnace where a mixture of argon and hydrogen flows.

The experimental setup for delayed hydride cracking on 1/4-in. thick compact tension samples has been constructed (Figure 3) and the generation of potential-drop calibration curves to monitor crack lengths during the experiments is currently being constructed.

<sup>1</sup> Idaho National Laboratory

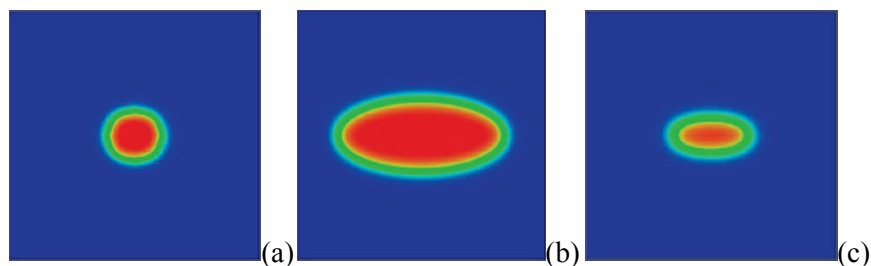


Figure 2. Modeling of hydride dissolution, with (a) initial seed at 500 K, (b) growth, and (c) dissolution of the grown precipitate when temperature is ramped to 800 K.



Figure 3. The experimental setup for delayed hydride cracking experiments. The furnace enclosing the sample and grip attachments is not shown in this figure.

## Benefits to DOE

The developments resulting from this project are also applicable to other hydriding alloy systems such as titanium and lithium; therefore, it will clearly provide a fundamental understanding of delayed hydride cracking.

## Publications

### ***Presentations***

Ahmed, K. E., P. Chakraborty, D. Schwen, S. Tysseyre, and S. B. Biner, 2016, “The effects of crack-tip stress fields on the delayed hydride cracking,” *Annual MST Meeting*, Salt Lake City, Utah, October 2016.

## Interns and Postdocs

Intern: Jordan Argyle

## 16-017—Evaluation of Load-Following Capabilities of Existing and New Nuclear Power Reactors in the Grid with Large-Scale Renewable Energy Sources

Haihua Zhao,<sup>1</sup> Hongbin Zhang,<sup>1</sup> and Curtis Smith<sup>1</sup>

### General Project Description

This project aims at evaluating readiness of operating and new reactors to have load-following capability facing the renewable energy challenge. Rapid development of renewable energy creates unique challenges for sustainability of nuclear power plants in some regions of the United States and around the world. The emerging early closure due to the extremely low (even negative) grid electricity price has proved that this is a sustainability issue for the existing power reactors. Early closures of a large number of nuclear power plants could impact electric system reliability and result in increased carbon emissions. This project directly addresses nuclear energy adaptation to grid dynamics, load following, and hybrid energy systems.

### Summary

During the first year of the project, we have focused our efforts on (1) understanding the problems of the load-following challenges from renewable energy; (2) identifying gaps and research areas; (3) developing an evaluation framework; and (4) building up capabilities for load-following evaluation for light water reactors (LWRs) and Generation IV reactors. In order to identify the major challenges for nuclear power plants in a grid with a large share of renewable energy, we performed a detailed literature review on load following capabilities and/or experiences, wind energy characters, methods to perform load-following for different reactors and integration methods for balancing renewable energy with nuclear power. The review covers both past U.S. experiences, ongoing Electric Power Research Institute flex operation research, Generation III and Generation III+ LWRs, small modular reactor work, and Generation IV reactors' capability for load following. European experiences, especially from France (i.e., too much nuclear energy) and Germany (i.e., too much renewable energy) were extensively reviewed. We also reviewed ongoing work on flexible operation of coal-fired power plants.

Major conclusions from review of the work include the following:

- Nuclear power reactors have inherent load-following capability with proper design, control, and monitoring, with some requiring moderate modifications. Nuclear reactors have better capability to perform load-following than other base-load thermal power plants (such as coal-fired power plants or natural-gas fired combined cycle power plants) in terms of power maneuver ranges or maintaining thermal efficiency at lower power.
- Extensive experiences exist in load-following of nuclear power plants in the past and in other countries, mainly in a pattern of slow and predictable ways.
- Even with the perception of an intermittent, unpredictable, and rapid change of power levels, the power variation rate in a grid with many wind turbines across a larger area can be quite limited and could be well within the capability range of nuclear power plants to perform load following.
- There is a lack of an integrated analytical evaluation model that would bring multiple disciplines together to provide risk-informed decision to allow flexible operations.

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<sup>1</sup> Idaho National Laboratory

We focused our efforts on establishing the integrated load following analysis capabilities at INL and developing a new methodology and new generation toolkit for load following, which could potentially release more margins than the current methods to better adapt reactors to the grid with a large amount of intermittent renewable energy. We identify five focused research areas to analyze and improve load following capabilities for existing or advanced reactors (Figure 1):

- Calculating and limiting local power peaking during rapid load changes
- Fuel performance analysis
- Advanced control methods for improving load-following capabilities
- Advanced power conversion systems' dynamic simulation and control for Generation IV reactors
- Risk and reliability analysis for key systems and components.

These five focused research areas leverage existing mature tools and advanced MOOSE-based tools. We have made significant progress in establishing evaluation capability, such as boiling water reactor core two-phase models, fuel performance analysis with both matured tool FRAPCON and advanced tool BISON, and advanced power conversion system dynamic simulation.

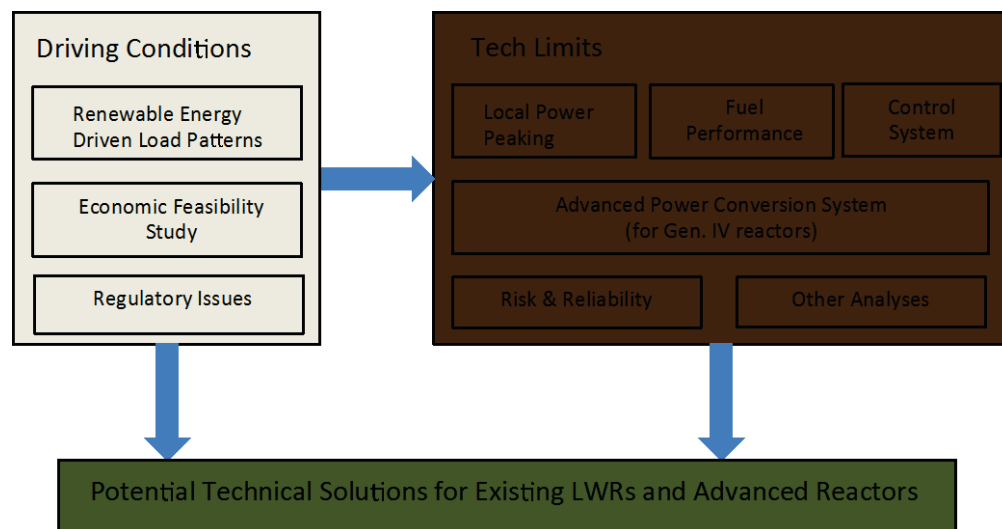


Figure 1. Evaluation framework for nuclear power reactors load following in the grid with a large scale of renewable energy sources.

## Benefits to DOE

This project helps address the sustainability issue of the current fleet's nuclear energy facing rapid development of renewable energy; therefore, it contributes to national energy security and diversity. The developed modern evaluation toolkit would improve the state-of-the-art of load following analysis and provide decision-makers with efficient and effective methods. The developed tool could be used for seeking further collaboration with nuclear vendors (both traditional and small modular reactor), U.S. Nuclear Regulatory Commission, INL hybrid energy system researchers, utilities, and state governments interested in developing energy markets. Because of the potential large impact on the nuclear industry, this research will enhance leadership of INL in nuclear energy.



## Publications

Zou, L., H. Zhao, and H. Zhang, 2016, “Newton-Krylov Method in Applications of Solving Two-phase Problems using Drift Flux Model,” *Proceedings of the ICAPP16*, San Francisco, California, April 17 through 20, 2016.

Zou, L., H. Zhao, and H. Zhang, 2016, “Solving Drift-flux Model Using High-Resolution Spatial Discretization Scheme,” *Transactions of the American Nuclear Society*, Vol. 114.

## Presentations

Zhao, H., H. Zhang, C. Smith, and S. Germain, 2016, “Evaluation of Load Following Capabilities of Existing and New Nuclear Power Reactors in the Grid with Large Scale of Renewable Energy Sources,” *LWRS RISMC and EPRI Meeting* at INL, August 2016.

## Interns and Postdocs

Interns: Yangmo Zhu, Guojun Hu, Lei Tu, and Jacob Ladd

## 16-026—Computationally Efficient Prediction of Containment Thermal Hydraulics Using Multi-Scale Simulation: Feasibility Study

Robert W. Youngblood,<sup>1</sup> N. T. Dinh,<sup>2</sup> and Igor Bolotnikov<sup>2</sup>

### General Project Description

The research is motivated by recognition that conventional high-resolution computational fluid dynamics (CFD) approaches are computationally overwhelming for simulation of the phenomenology of accident scenarios in reactor containment, because these scenarios evolve over long time scales and extend over large length scales. The difficulties are even more pronounced when a sensitivity/uncertainty analysis is required to support risk-informed design and safety analysis of nuclear power plants, because this entails simulation of many time histories in order to develop an understanding of the implications of the variabilities and uncertainties that affect the evolution of the scenarios. The project's technical objective is to develop a technical basis for a coarse-grained CFD capability that is needed for high-fidelity analysis of containment thermal-hydraulics. Specifically, the project investigates a data-driven, multi-scale framework having the potential to enable computationally efficient simulation of containment thermal-hydraulic processes.

### Summary

The work is being carried out in two parallel threads by two doctoral candidates, Han Bao and Botros Hanna. Han Bao's study focused on demonstration of a mesh optimization system in thermal-hydraulic simulation. This work comprised several developments and case studies. First, a case study was done illustrating the sensitivity of key results on meshing. The first study involved suppression pool stratification; spatial averaging of suppression pool temperature gives a seriously misleading picture of the net positive suction head available to RCIC (i.e., Reactor Core Isolation Cooling), and correspondingly incorrectly predicts the time of RCIC failure. The next case study involved nodalization in the core region. It was shown that both the peak values and the time dependence of peak cladding temperature vary considerably with nodalization. These results are not a surprise, but simply point out the need to understand the effects of nodalization. The overall project's aim is to learn how to compensate for the effects of coarse mesh in a simulation that still executes efficiently. Accordingly, Han Bao embarked on development of the optimized mesh/model information system. The goal is to develop the optimized mesh/model information system as a smart data-driven system for advising on optimized selection of mesh size and model for simulation of new physically similar conditions using a coarse-mesh code to achieve higher accuracy. The principles were illustrated by numerical experiments: (1) one-dimensional heat conduction simulation, where Han first solved the problem with high fidelity, and then showed how to formulate and train a much cruder model to give essentially the same result (small error in the crude-model result); and (2) two-dimensional natural convection simulation. A somewhat analogous process was carried out for the problem of natural convection in a volumetrically heated fluid with a cold-wall boundary condition at the top of the volume and an isolated bottom wall. The system was modeled in the Westinghouse simulation code GOTHIC, with different meshes and model parameters. An optimal mesh was determined and it was shown that the error in the resulting cruder model is small.

The study demonstrated the feasibility of a physics-based, data-driven mesh-optimization system for a system-level thermal-hydraulic simulation, which aims to advise on the optimized selection of mesh size and model for coarse-mesh codes with low-fidelity physical models to achieve higher accuracy via a machine learning method. The theoretical basis and assumptions have been discussed for development and assessment of the optimized mesh/model information system model. A Gaussian process regression method has been applied as the

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<sup>1</sup> Idaho National Laboratory

<sup>2</sup> North Carolina State University

machine learning tool for data training. Two case studies, including a conduction and natural convection problem, have been completed and analyzed. More theoretical work, especially on error estimation and uncertainty quantification based on the machine learning method, is proposed for FY 2017.

Botros Hanna's work is in the same vein (i.e., learning how to develop simplified models informed by experimental data and high-resolution models), but Botros's work focuses explicitly on coarse-grained CFD and has a multi-physics aspect. Analysis of the turbulent natural convection case with volumetric heating in a horizontal fluid layer is performed with a high-fidelity simulation (with fine mesh) and lower-fidelity, three-dimensional simulation (coarse mesh). The coarse mesh simulation was corrected by modifying the diffusion term in the energy equation. This global correction was presented as a function of mesh size. The quasi-steady-state temperature distribution predicted with a coarse mesh is corrected without capturing the initial transient. We hypothesize that the same correction can be applied to different cases having the same Rayleigh number. "Learning" this term (i.e., in the machine-learning sense) enabled the coarse-mesh model to emulate the fine-mesh model successfully.

The work summarized above demonstrated an encouraging level of success in teaching low-resolution models to emulate high-resolution models. This represents meaningful progress toward an ambitious goal.

## Benefits to DOE

The need to be able to analyze the phenomenology of reactor accidents has been recognized for a long time and re-emphasized by events at Fukushima. Current state-of-practice capabilities for doing this involve lumped-parameter system codes. These system codes have improved over time, but their remaining acknowledged weaknesses for simulation of phenomena occurring over large regions and for long times leave the need to be able to analyze phenomena in containment unmet. The relevance of this work is high.

Moreover, although presently motivated by simulation and analysis of containment thermal-hydraulic processes, the insights derived from the project and the data-driven multiscale simulation methodology that is being developed are applicable to other complex thermal-hydraulic processes needed for advanced energy system safety analysis.

## Publications

- Bao, H., N. Dinh, H. Zhao, H. Zhang, L. Zou, and R. Szilard, 2015, "Simulation of BWR Mark I Station Black-Out Accident Using GOTHIC: An Initial Demonstration," *ANS Winter Meeting*, Washington D.C., November 8 through 12, 2015.
- Bao, H., N. Dinh, H. Zhao, H. Zhang, L. Zou, and R. Szilard, 2016, "A Study of BWR Mark I Station Blackout Accident: GOTHIC Modeling and Sensitivity Analysis," *International Congress of Advanced Nuclear Power Plants*, San Francisco, California, April 2016.
- Hanna, B., N. Dinh, and I. Bolotnov, 2016, "A Framework for Data-Driven Model Development for Coarse-Grained Computational Fluid Dynamics," *International Topical Meeting on Advances in Thermal Hydraulics 2016 (ATH 16)*, New Orleans, Louisiana, June 12 through 16, 2016.
- Lin, L., N. Dinh, R. Sampath, and N. Akinici, 2016, "A Computational Study of Thin Film Dynamics on Micro-Structured Surfaces," *Proceedings of the ASME 2016 Summer Heat Transfer Conference*, HT2016-7382, Washington D.C., July 10 through 14, 2016.
- Lin, L., 2016, "Assessment of the Smoothed Particle Hydrodynamics Method for Nuclear Thermal-Hydraulic Applications," Master of Science Thesis in Nuclear Engineering, North Carolina State University, 75 p, April 2016.

## **Interns and Postdocs**

Interns: Han Bao, Yangmo Zhu, and Linyu Lin

## **Collaborations**

University: North Carolina State University

## 16-033—Investigation of Gadolinium Nanocrystal Gels for Scintillator Use in Neutron Detection

Catherine Riddle<sup>1</sup> and Douglas Akers<sup>1</sup>

### General Project Description

Development of new neutron detectors to replace helium-3 ( $^3\text{He}$ ) detectors is imperative due to a worldwide shortage of  $^3\text{He}$  following the drawdown in nuclear weapons production since the end of the Cold War. The U.S. Department of Homeland Security would like to deploy monitors for detection of neutron emissions from shipping containers housing illicit nuclear material; however, this effort has been put on hold until new replacements for  $^3\text{He}$  detectors can be developed. The investigation of a unique, first-of-its-kind, gadolinium (Gd)-loaded nanocrystal gel scintillator has led to development of the next generation of neutron detection material for the replacement of  $^3\text{He}$ . This new gel material, SHINE (i.e., scintillation hydro-gel for isotopic neutron emitters), uses water soluble Gd and wavelength tunable nanocrystals in a gel scintillator. This novel detection material incorporates the best properties of liquid and solid scintillators, without the disadvantages such as continuous filtering to keep liquids free of contaminants, slow throughput of containers, higher base component costs, ‘dead’ voids in solid scintillators, and lower overall efficiencies than Gd (Dijkstra et al. 2011).

### Summary

SHINE was developed using a unique combination of polyacrylamide, water soluble Gd-diethylenetriaminepentaacetic acid (Gd-DTPA), and a new fluorescent nanocrystal, known as quantum dots (QDs). The QDs consist of water-soluble zinc sulfide (ZnS) or zinc oxide (ZnO) core shells (shown in Figure 1 as the incorporated gel, which can fluoresce in the presence of energy signature (such as gamma rays) (Gao et al. 2010, Li et al. 2007, Mandal et al. 2012). This novel scintillator exploits the positive aspects of Gd, which has the highest thermal neutron capture cross section of any natural element, and highlight yield fluorescent QDs in a unique, water-based, environmentally friendly gel that can be poured into any shape or size and solidified within minutes at room temperature. SHINE has been successfully tested for neutron detection and shows promise as both a replacement for current  $^3\text{He}$  neutron detectors and potential use in hand-held, compact neutron detection units and antineutrino detection for nuclear reactor safety and the International Atomic Energy Agency. SHINE’s sister technology, SHADE (i.e., scintillation hydro-gel for antineutrino detection) will soon be tested at INLs ATR facility. SHADE was developed alongside SHINE and shares similar properties with its sister technology as an environmentally friendly alternative to toxic compounds used in antineutrino detection for nuclear reactors. The Gd in SHINE/SHADE showed an excellent response to neutrons from an americium-beryllium source (Figure 2), given that a series of standard peaks is representative of neutron absorption and gamma ray emission. The Gd loading for SHINE is significantly higher (i.e., less than or equal to 15% Gd) than what is achievable in current commercial detectors that can only load about 3% Gd in crystal, plastic, or water scintillators.

A first-of-its-kind scintillation gel/nanocrystal detection material has been synthesized in a one-step process at room temperature. The SHINE material forms a stronger and more durable gel structure in the presence of Gd than in the polyacrylamide alone. The Gd-DTPA is extremely soluble in water and remained homogeneous throughout the gelling process, forming an optically transparent, rigid gel with high Gd concentrations. The Gd-DTPA gel shows a good detection response for neutron conversion to gamma rays. The water soluble ZnS core-shell QDs performed well with respect to gel synthesis and optical transparency in the presence of Gd-DTPA. However, an increase in the QD concentration is needed in order to increase light yield to a level comparable to, and even higher than, current sodium iodide detectors (Zmeskal 2015). With the ability to produce higher concentrations of ZnS QDs, SHINE will meet its full potential as the next generation in neutron detection

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material. The investigation of Gd nanocrystal gels has yielded a novel scintillation material in the form of SHINE and additional funding has been established in order to optimize the light yield of the gel so that it may be commercially licensed and be nominated for a Research and Development 100 Award for 2017.

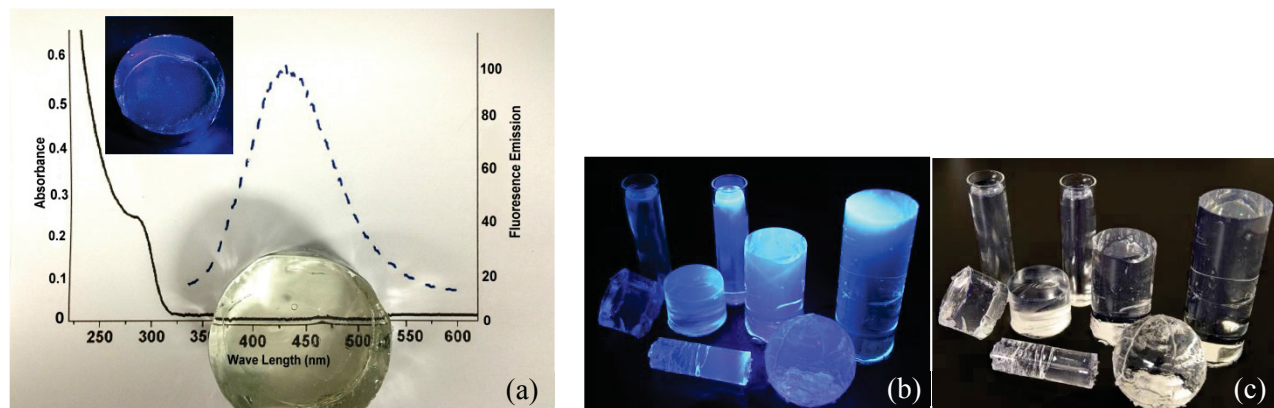


Figure 1. (a) ZnS and CdSe/ZnS QDs in a Gd-DTPA/polymer gel showing optical transparency and emission wavelength. Inset on (a) and (b), SHINE gel under ultraviolet light and (c) SHINE in ambient light.

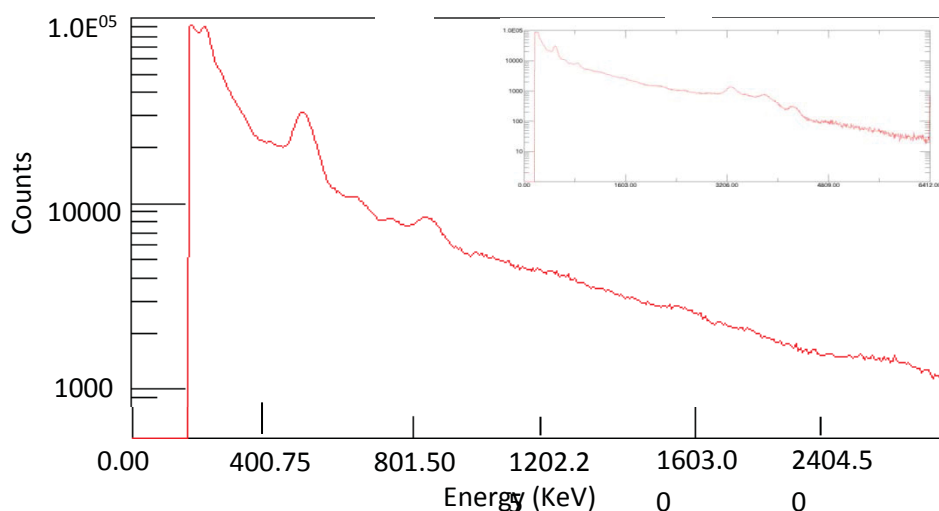


Figure 2. Gd-DTPA gel (i.e., less than 15% Gd loading) response to an americium-beryllium source. Full spectra, inset.

## Benefits to DOE

This research will reinforce the role of INL as an innovator for real-world problems. This LDRD work has found a novel solution to a current real-life problem affecting the world and it has the potential to be a game changer as the next generation of neutron and antineutrino detection material. SHINE has a patent pending and a strong opportunity for industrial partnerships for the replacement of current  $^3\text{He}$  neutron detectors in the war against nuclear subversion. Furthermore, this project has reinforced the role of INL as a key player in the National Nuclear Security Administration arena and increased the visibility of INL in the Department of National and Homeland Security, expanding studies into other areas of nuclear safety and nonproliferation.



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## Invention Disclosures, Patents, Copyrights

- U.S. Patent 9,505,977 B2, Riddle et al., “Gadolinium-Loaded Gel Scintillators for Neutron and Antineutrino Detection,” issued November 29, 2016.

## 16-036—Neutron Microscope to Enable High-Resolution Neutron Tomography at INL

Muhammad Abir,<sup>1</sup> Walter J. Williams,<sup>1</sup> and Boris Khaykovich<sup>1</sup>

### General Project Description

Neutron radiography is an important tool for post-irradiation examination of materials. At INL, the neutron radiography facility is used for examining fuel elements. Usually, neutron radiography instruments are designed as pinhole cameras, where a neutron beam from a small aperture (i.e., pinhole), with a diameter  $D$ , illuminates a sample at a large distance  $L$ , where  $L/D$  is usually between 50 and 500. The spatial resolution of such a system improves with an increasing  $L/D$  ratio and with a decreasing sample-to-detector distance. The high  $L/D$  ratio restricts the neutron flux on the sample, sometimes severely, by restricting the solid angle of the source viewed from the sample position. Where powerful optical tools (such as focusing mirrors) are available for neutron radiography, they might bring transformative improvements by increasing the spatial and temporal resolution of neutron instruments by orders of magnitude. The purpose of this project is to design and implement a “proof-of-concept” for these focusing mirrors, sometimes called Wolter mirrors, for the neutron radiography application at the Neutron Radiography Reactor (NRAD) at INL. The objective is to test how well this would work as a magnification-one neutron microscope, where Wolter mirrors are placed between the sample and the detector and work as image-forming optics. The spatial resolution of this system does not depend on the  $L/D$  ratio, allowing illumination of the sample from a large-diameter source. In addition, the large separation between the radioactive spent fuel sample and the detector would shield electronic detectors from damage by gamma rays.

### Summary

The university collaborators at the Massachusetts Institute of Technology (MIT) are leading the design of the neutron focusing optics for potential manufacturing and installation at INL. INL established active collaboration with MIT and the MIT group is instructing the principal investigator on the details of Wolter mirrors for the microscope and ray-tracing simulation methods and software, which have been developed at MIT. At the same time, the MIT group is learning the details of the INL neutron imaging setup at NRAD.

We have identified the candidate geometrical configurations of the mirrors. The mirrors are shown schematically in Figure 1; for clarity, only two reflections are shown, the paraboloid-paraboloid geometry.

For NRAD, each paraboloid will be augmented by confocal hyperboloid, resulting in neutron trajectories having four reflections. The reason for the four reflections is the thermal-neutron spectrum at NRAD, resulting in small critical reflection angles. In order to be reflected by the mirrors, neutrons have to approach the mirror surfaces at the angle below the critical angle, which is rather small, for thermal neutrons of  $\lambda=2\text{\AA}$ , the critical angle is only about  $0.2^\circ$  for Ni. The optics will consist of two identical pairs of mirrors forming a system with magnification one. This design will achieve the highest throughput (i.e., signal rate at the detector). Our analysis showed that an increase of the neutron flux at the detector of two orders of magnitude is possible when compared with the existing system. The combination of a significant increase in flux, with the possibility of using a digital detector, would allow fully automatic tomographic measurements at NRAD, which is a quantitatively different approach that would enable new science.

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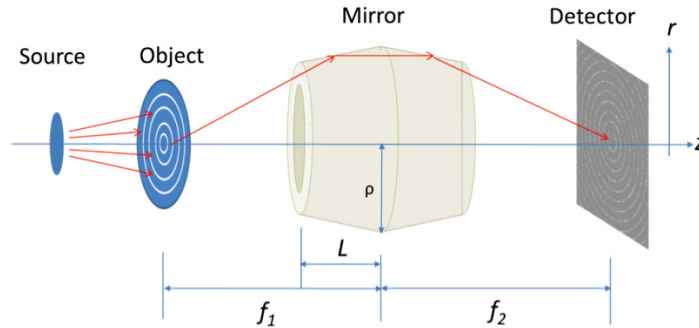


Figure 1. Schematic illustrations of a neutron imaging system with a test object used to calculate resolution. The object (i.e., sample) and the detector are placed at the focal planes of the focusing mirrors. The optics of magnification one consist of two identical mirrors, each made of confocal hyperboloid and paraboloid. For simplicity, only paraboloids are drawn here. Approximate sizes are  $f_1 = f_2 \sim 2$  m,  $L \sim 0.1$  m,  $\rho \sim 0.03$  m.

In order to increase the signal rate further, the mirrors for the INL facility will have to be coated with Ni/Ti multi-layers (i.e., the so-called neutron supermirrors). The supermirrors have the critical reflection angle that is higher than that of Ni, which possesses the highest critical angle of all natural elements. The supermirrors are produced by sputtering of Ni and Ti layers with thickness designed according to standard algorithms. Although flat neutron supermirrors are commercially available, Wolter supermirrors with the critical angle twice that of Ni have been recently made. In fact, the first test of Wolter supermirrors in the neutron beam has taken place just days ahead of this report at the National Institute of Standards and Technology Center for Neutron Research. The reflectivity of the supermirrors has been tested by imaging a small source and measuring the integrated intensity of the image as a function of neutron wavelength. Because Wolter supermirrors would be necessary for the INL facility, the MIT group has simulated the expected intensity and is working on the data analysis from that test. The intensity was simulated using the known reflectivities of flat neutron supermirrors of the same structure. Figure 2 shows the simulated intensity as a function of the wavelength. The sudden increase in the intensity at about 3 Å is due to reaching the critical angle. Note that the mirrors used in this test were not yet optimized for NRAD.

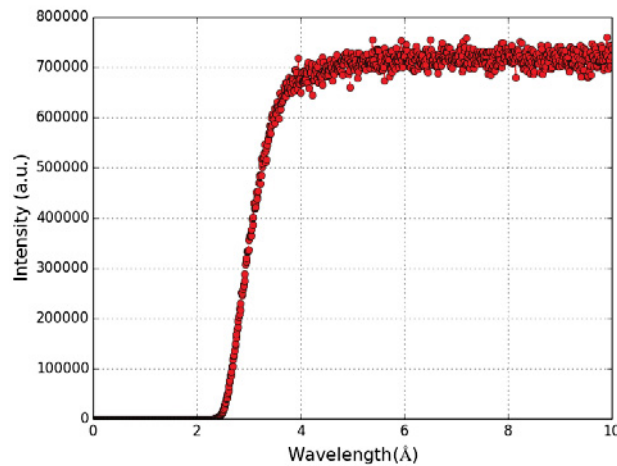


Figure 2. Simulated dependence of the intensity of the focal spot as a function of neutron wavelength. The mirror surface is coated with an Ni/Ti multilayer. The mirrors are small test mirrors not optimized for NRAD. However, similar coating will be prepared for mirrors optimized for NRAD. The edge at 3 Å results from

*neutrons reaching the critical reflection angle. The exact position of the edge will depend on the mirrors' geometry. For the four-reflection system, the edge would shift toward about 1.5 Å.*

Another important consideration for grazing-incidence optics is the field-curvature aberrations. We are developing compensation solutions for the aberrations, based on either curved detector or coded-aperture designs.

## Benefits to DOE

The project would benefit DOE by allowing a new post-irradiation examination method to study spent nuclear fuel. Fully automatic neutron tomography would allow a clear understanding of the mechanical stability of the irradiated fuel elements with high spatial resolution. The need for such investigations is clear, because new fuel has been developed for existing and new nuclear reactors. The innovation of this project is in replacing 500-year-old technology of pinhole imaging with modern focusing optics. This project fosters close collaboration between MIT and INL, including training of students and postdocs.

## Publications

Abir, M. and B. Khaykovich, 2016, “Neutron Focusing Mirrors to Enable High Resolution Neutron Imaging at Idaho National Laboratory,” *Physics Procedia*, in preparation.

## Presentations

Abir, M., 2016, “Neutron Focusing Mirrors to Enable High Resolution Neutron Imaging at Idaho National Laboratory,” oral presentation, *ITMNR-8*, Beijing, China, September 6, 2016.

Khaykovich, B., 2016, “Neutron Focusing and Guiding Using Multilayer-Coated Nickel Mirrors,” oral presentation, *American Conference on Neutron Scattering*, Los Angeles, California, July 12, 2016.

## Interns and Postdocs

Interns: Huarui Wu and Jonathan Morrell

Postdocs: Durgesh Rai and Muhammad Abir

## Collaborations

University: Massachusetts Institute of Technology

## 16-040—Integration of Prognostic Techniques and Probabilistic Safety Assessment for Online Risk Monitoring

Vivek Agarwal,<sup>1</sup> Andrei V. Gribok,<sup>1</sup> and Curtis L. Smith<sup>1</sup>

### General Project Description

Nuclear power plants use risk monitors to estimate system risk for ensuring safe and reliable operation of the plant. The existing risk monitors, based on probabilistic risk assessment (PRA), provide system risk based on the current plant configuration. Even though traditional PRA seeks realistic results, the assessment is based on Boolean logic (i.e., components have only two states: operational and failure), time-independent failure information (i.e., failure information is collected in time snapshots and not on a continuous basis), and assumptions that system/components are non-repairable. As a result, traditional PRA in its present form has some limitations: (1) it is not capable of handling time-evolving scenarios; (2) it does not include system/component degradation or aging information; (3) it is a conservative risk estimate; and (4) it is unable to handle uncertainty due to changes in reliability of components/system as a result of operational or external factors.

This research is focused on developing an enabling approach to make strides in current risk monitors to provide time and condition-dependent risk. The approach adapted in this research is integrating traditional PRA models with condition monitoring and prognostic techniques (Figure 1). Mathematically, it can be interpreted as a transition from  $\{0,1\}$  to  $[0,1]$ . In  $\{0,1\}$  only Boolean states are considered, whereas in  $[0,1]$  all possible states between 0 and 1 (including 0 and 1) are considered.

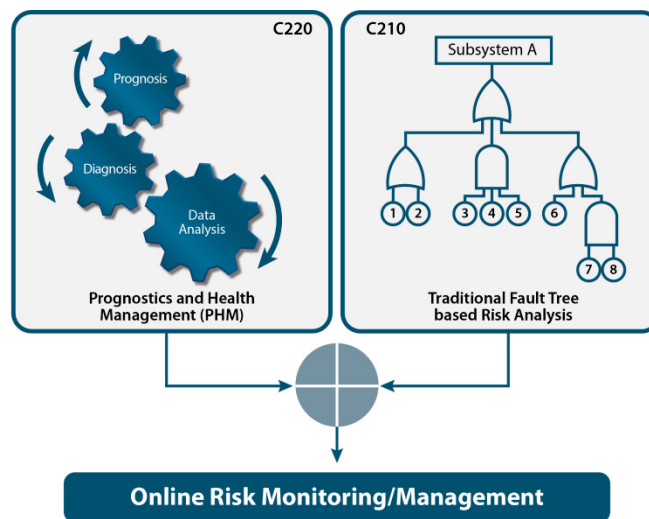


Figure 1. Enhancing traditional risk analysis with prognostics and health management.

### Summary

The first year of this 3-year project focused on understanding and identifying key assumptions that would be required to achieve online risk monitoring, followed by collecting measurement data, including historical failure data across the current fleet of light water reactors. First year highlights are as follows:

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- Component importance measures are widely used in tandem with PRA to prioritize components for purposes of decision making such as maintenance planning. The literature review activity identified and applied several key importance measures widely used in the nuclear industry. Measures were identified that could be extended to repairable systems with non-binary states (Van de Borst and Schoonakker 2001).
- With too much emphasis on risk and safety significance importance measures, little attention was paid to the cost incurred to maintain systems and their components within a given time period. This led to investigation of cost-based importance measures. Three types of costs are identified to account for the cost-based importance measure of components: (1) cost of improving component reliability, (2) cost due to component failure, and (3) cost of system failure. The existing research only includes the cost of component failure and cost of system failure for calculating cost-based importance measures (Wu and Coolen 2013). As part of this research, the cost-based importance measure calculation was extended to incorporate all three costs to include the valuable cost of improving component reliability of a repairable system.
- The measurement and failure data for different components were obtained from the reactor operation experience results and databases made available by the U.S. Nuclear Regulatory Commission and from the risk monitoring data of ATR. The data from the U.S. Nuclear Regulatory Commission (Eide et al. 2007) characterize industry-average performance for components and initiating events at U.S. commercial nuclear power plants. Collaborative efforts have been initiated with the risk monitoring team at ATR. Future efforts will focus on using the U.S. Nuclear Regulatory Commission data and ATR data to better understand component aging and degradation in nuclear power plants operating under various conditions.
- In order to transform the binary nature of traditional PRA to a continuous form, a special class of real-valued functions, called Rvachev functions (R-functions), was studied. Research to represent a Boolean logic through continuous differential function describing Venn and Euler's diagrams of the system using R-function is in progress.

## Benefits to DOE

This research will directly benefit work being conducted under the Light Water Reactor Sustainability Program by enhancing existing PRA capabilities to incorporate component aging and degradation. This research work facilitates and promotes inter-departmental collaboration at INL through an active partnership between the two departments of Human Factors, Controls, and Statistics and Risk Assessment and Management Services within the Nuclear Science and Technology Directorate. This research led to staff growth from three to four in the Human Factors, Controls, and Statistics department by hiring one postdoctoral researcher. Additionally, two interns were also hired as part of this project.

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## Publications

- Agarwal, V., P. Chatterjee, A. V. Gribok, and C. Smith, 2016, "Online Risk Monitoring of Nuclear Power Plants: System and its Components Ordering Based on Cost Importance," in *Proceedings of the 13<sup>th</sup> International Conference on Probabilistic Safety Assessment and Management*, Seoul, S. Korea, October 2 through 7, 2016.



## **Interns and Postdocs**

Interns: Payel Chatterjee and Brittany Layne Umbrage

Postdoc: Vaibhav Yadav

## 16-046—Development of a Synergistic Approach to Study Irradiated Materials Using Coupled Experiments and Simulation

Assel Aitkaliyeva,<sup>1</sup> Dan Wachs,<sup>1</sup> Cynthia Papesch,<sup>1</sup> and Michael Tonks<sup>2</sup>

### General Project Description

Understanding and predicting the effects of irradiation damage on material properties is possible only when state-of-the-art experimental characterization methods are used in conjunction with advanced mesoscale modeling and simulation. For this reason, DOE and INL have invested heavily in facilities and capabilities for microstructure characterization of irradiated materials, as well as development of the MOOSE/BISON/MARMOT code system for predicting microstructure evolution and its impact on properties in fuel and cladding materials. This project shifts the focus from building capabilities to demonstrating the use and integration of these capabilities to shorten the nuclear fuel development cycle. To fully realize the benefits of tightly coupled experiments and simulation at the microstructural level, this project works in the following three areas: (1) developing experimental procedures to obtain the specific pre and post-irradiation characterization data required for validation and uncertainty quantification of MARMOT models, (2) demonstrating the value of a coupled experimental and simulation approach on understanding critical thermal properties in a material of broad interest, and (3) understanding the evolution of microstructure under transient irradiation conditions and its impact on properties for use with the Transient Reactor Test Facility (TREAT).

### Summary

The project is conducting systematic microstructural and thermal property characterization, along with multi-scale modeling of nuclear fuels before and after irradiation. This characterization and modeling will provide much-needed linkage between thermal properties and microscopic-scale observation. To achieve such characterization, a combination of complementary techniques, such as scanning transmission electron microscopy/transmission electron microscopy (TEM) and others, was implemented. Previously, preparation of transuranic fuels for transmission electron microscopy has been associated with a number of challenges and deemed impossible. Recent utilization of focused ion beam instruments in the nuclear fuels field helped overcome these issues. Microstructural characterization of the cast fuel, which includes analysis of the structure and chemistry of the primary, secondary, and impurity phases, and lattice defects, has been conducted and representative results obtained for one of the alloys selected for this study are shown in Figure 1.

Microstructural characterization was linked to thermal properties analysis. Measurement of thermal properties, which are focused on thermal conductivity, phase transition temperatures, and enthalpy of the phase transitions, has been conducted on selected fuel alloys. Thermal properties of the materials were measured using a variety of instruments. Transition temperatures, enthalpies of transition, and heat capacities were determined employing a differential scanning calorimeter/thermogravimetric analyzer. Thermal diffusivities and conductivities of the fuels were determined using laser flash analyzers. Phase transitions observed during differential scanning calorimeter measurements were correlated with the phases observed in TEM.

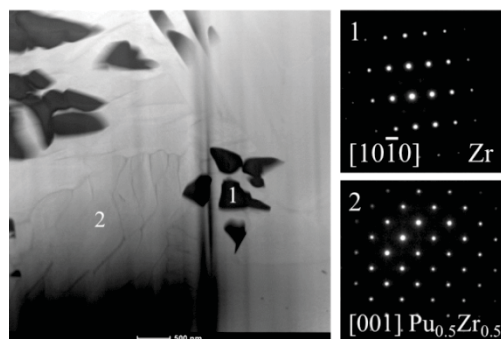
This correlation information is being used as input to the MARMOT code and will result in precise modeling of the microstructural features that impact the thermal conductivity of the fuel. A validated simulation of the unirradiated alloy will be used as a base for modeling of the fuel microstructure evolution upon irradiation. It will also develop new capabilities in MARMOT for using experimental data for uncertainty quantification and for modeling U-Pu-Zr fuel in the next fiscal year. Finally, TREAT will be implemented as a science tool for attaining

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<sup>1</sup> Idaho National Laboratory

<sup>2</sup> Pennsylvania State University

in-depth understanding of metal fuel evolution upon irradiation and to further promote the application of MARMOT to transient conditions.



*Figure 1. Scanning transmission electron micrographs of the phases observed in Pu-30Zr fuel and corresponding selective area diffraction patterns. The microstructural information is correlated with thermal properties and used as an input to MARMOT.*

Acquisition of the specific property data will allow precise modeling of the microstructural features that impact thermal conductivity and provide a validation simulation of the base, unirradiated fuel alloy. The experimentally determined phases are now being used for direct reconstruction of the fuel microstructure in MARMOT. Volume fractions of each phase have been determined from TEM micrographs and are now used as an input parameter for the MARMOT. The detailed procedure outlining best practices for collecting data specifically targeted for mesoscale model development has been written and is now being implemented for each analyzed sample.

During the first stage of this project, characterization and modeling of unirradiated fuel has been conducted. In FY 2017, fuel alloys will be irradiated in TREAT to induce a succession of microstructural changes (e.g., grain growth, phase transformation, and thermal-mechanical stresses) under well-defined irradiation conditions. These fuel alloys will be recharacterized after irradiation in FY 2018 to provide the data required to evaluate analytical predictions of the new transient capability in MARMOT. The specific irradiation conditions are currently being established based on preliminary MARMOT calculations.

## Benefits to DOE

This work will provide established procedures for coupling experiments with modeling and simulation to investigate critical material behavior in nuclear fuel. This coupled approach has the potential to provide detailed and quantitative understanding of atomic scale in-reactor degradation behavior of nuclear fuels. This understanding is of critical importance to development of next generation reactor systems, because evolution of the microstructure at the atomic scale has the most profound impact on bulk properties and in-reactor performance. These procedures will include application of various complementary techniques to provide a detailed characterization of the fuel before and after irradiation. As a result of this effort, DOE will have the kind of information needed to provide a more thorough understanding of radiation response of metallic fuels and shed light on its future as a fuel for next generation fast reactors.

## Publications

Aitkaliyeva A. and C. Papesch, 2016, "Identification of Subsurface Phases in Ternary U-Pu-Zr Fuel," *Journal of Nuclear Materials*, Vol. 473, pp. 75.

### ***Presentations***

Aitkaliyeva, A. and C. Papesch, 2016, “TEM Identification of Phases in Metallic Pu-Based Fuels,” *2016 Microscopy and Microanalysis Conference*, Columbus, Ohio, July 2016.

Aitkaliyeva, A., J. Madden, and C. Papesch, 2016, “TEM Investigation of Phases Formed in Ternary U-Pu-Zr Systems,” *2016 TMS Annual Meeting & Exhibit*, Nashville, Tennessee, February 2016.

### **Collaborations**

University: Pennsylvania State University

## 16-050—Stress Corrosion Cracking Testing in Supercritical Carbon Dioxide

Sebastien Teyssyre,<sup>1</sup> Julie Tucker,<sup>2</sup> Piyush Sabharwall,<sup>1</sup> Joe Palmer,<sup>1</sup> and Robert Fox<sup>1</sup>

### General Project Description

This project is developing testing capability to address stress corrosion cracking issues in supercritical carbon dioxide. In addition, this project is generating corrosion data. This work aims to help select structural materials for supercritical carbon dioxide energy systems.

### Summary

In FY 2015, a corrosion loop was built at Oregon State University. This loop is used to generate corrosion data on nickel-based alloys (e.g., Inconel 625 and Haynes 282) and their joints (i.e., diffusion bonding, laser welding, and brazes) at temperatures ranging from 300 to 750°C. In FY16, in addition to analyzing corrosion data (e.g., nature of the oxide and degradation of the underlying material), the project team is working on adapting a technology that was developed at INL for stress corrosion cracking testing in a nuclear reactor to the corrosion loop. This work will allow the project team to perform the first in line measurement of stress corrosion cracking crack growth in supercritical carbon dioxide.

The characterization of corrosion in supercritical carbon dioxide (CO<sub>2</sub>) led to the following information:

- At 750°C lead to significant void density underneath the oxide layer and internal oxidation. A high concentration of aluminum along the oxidized grain boundary has been observed. The primary surface oxide was identified as Cr.
- At 650°C, oxide pegs or protrusions growing into the matrix were shown to have a greater depth and density with longer exposure times.
- At 650°C, there was noticeable evidence of precipitation at grain boundaries and intragranularly; however, underneath the scale, there was a precipitate-free zone that is also associated with a Cr depletion area and a layer of newly formed, possibly recrystallized grains. The as-received sample has no recrystallized zone compared to the cross-sectional results seen after 200 and 600 hours.
- At 650°C at various time intervals, the recrystallization zone underneath the oxide was observed.
- The mean thickness of scale increased with a function of temperature but stayed relatively similar as a function of time at 650°C. The precipitate-free zone grew slightly as a function of time.

In regard to implementation of stress corrosion cracking testing capability, the implementation of the stress corrosion cracking monitoring and control in existing corrosion set ups has been performed. The requirements to perform stress corrosion cracking in this environment (i.e., high temperature and pressure) were investigated and it was confirmed that the bellow system we are considering will be appropriate. The experiment control system was designed and the components were purchased.

### Benefits to DOE

S-CO<sub>2</sub> Brayton conversion will benefit efforts in the Offices of Nuclear Energy, Fossil Energy, and Energy Efficiency and Renewable Energy. To be able to deploy such technology, it is necessary to select suitable materials to be used for the components working in this environment. This work develops stress corrosion

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<sup>2</sup> Oregon State University

cracking testing capability to generate the first stress corrosion cracking data in supercritical CO<sub>2</sub> and support corrosion experiments.

This work equips INL and its partner with the equipment and knowledge for addressing stress corrosion cracking and its mitigation for supercritical CO<sub>2</sub> systems. Vito Cedro (National Energy Technology Laboratory project manager) already expressed interest in this work and its future development.

## Publications

### ***Presentations***

Teeter, L., F. Teng, W. Marcum, J. Kruzic, M. Anderson, and J. D. Tucker, 2015, “Supercritical Carbon-Dioxide System for Materials Corrosion Testing,” *Oregon State University Engineering Research Expo*, March 4, 2015.

Teeter, L., F. Teng, W. Marcum, J. Kruzic, M. Anderson, and J. D. Tucker, 2015, “Supercritical Carbon-Dioxide System for Materials Corrosion Testing,” *TMS Annual Meeting and Exhibition*, Orlando, Florida, March 15 through 19, 2015.

Teeter L., B. Adam, W. Marcum, J. Kruzic, M. Anderson, and J. D. Tucker, 2015, “OSU Supercritical Carbon-Dioxide System for Materials Corrosion Testing,” Idaho National Laboratory, April 20, 2015, invited.

Teeter L., B. Adam, W. Marcum, J. Kruzic, M. Anderson, and J. D. Tucker, 2015, “Corrosion of Energy System Materials in Supercritical Carbon Dioxide (SC-CO<sub>2</sub>),” *EPRI International Conference on Corrosion in Power Plants*, San Diego, California, October 13, 2015.

Teeter, L., N. Huerta, O. Dogan, M. Ziomek-Moroz, R. Oleksak, D. Oryshchyn, C. Disenhof, J. Baltrus, and J. Tucker, 2016, “Corrosion Behavior of Austenitic Stainless Steel in Supercritical CO<sub>2</sub> Containing O<sub>2</sub> and H<sub>2</sub>O,” *229<sup>th</sup> Electrochemical Society Meeting*, San Diego, California, May 29 through June 2, 2016.

## Interns and Postdocs

Interns: Lucas Teeter, Allison Burns, Patrick Jarrold, Ben Adams, and Reyixiati Repukaiti

## Collaborations

University: Oregon State University



## 16-055—Capability Extension for Multi-Scale, Multi-Application Development within the Multi-Physics Object-Oriented Simulation Environment

Cody Permann,<sup>1</sup> David Andrs,<sup>1</sup> Derek Gaston,<sup>1</sup> John Peterson,<sup>1</sup> Andrew Slaughter,<sup>1</sup> and Brian Alger<sup>1</sup>

### General Project Description

The Multi-Physics Object-Oriented Simulation Environment (MOOSE) is INL's preferred modeling and simulation framework. It is the underlying framework for several mature research and development codes, including BISON (Williamson et al. 2012) and Marmot (Tonks et al. 2012). Additionally, MOOSE is used by private industry and as a simulation platform at other laboratories and universities internationally. The purpose of this project is to support development and implementation of several experimental algorithms within the framework, enabling forward-thinking analysis and one-of-a-kind capabilities for challenging problems facing the numerical simulation and modeling community. New and developing technologies will be implemented and integrated for use by all codes and programs built on the MOOSE framework.

### Summary

Three separate tasks were undertaken during the first year of this project: (1) development and deployment of a customized continuous integration tool, (2) implementation and analysis of an adaptive Runge-Kutta time integration capability, and (3) prototype implementation of a reconstructed discontinuous Galerkin (rDG) method. Each of these is described separately.

A new tool (i.e., the continuous integration, verification, enhancement, and testing [CIVET] application) was developed using the experience gained from our pilot "MooseBuild" project from 2015. CIVET is a highly flexible, stand-alone, client/server application that allows robust continuous integration and testing, complete with a dashboard and web interface. CIVET stands apart from the plethora of open-source continuous integration tools because it maintains revision control for *both* the "recipes" it uses and the test results it produces, enabling it to meet the stringent NQA-1 requirements for software quality. The capabilities of the CIVET application were reported as "notable practices" by Nancy Kyle and Michael Lackner, external NQA-1 assessors. CIVET also features a distributed continuous integration model, where developers at different sites can run their own clients and report local results to a master results server. CIVET has been approved for release as open-source software pending final legal approval.

The adaptive Runge-Kutta time integration task was broken into two major tasks: (1) implementation of the Time Integrator objects in MOOSE underlying the scheme and (2) implementation of an adaptive time stepper object in MOOSE. Part 1 was completed in 2016. The following explicit Runge-Kutta time integrators are now available in MOOSE: Ralston's method, the explicit midpoint method, and Heun's method, also known as the second-order total variation diminishing Runge-Kutta method. In addition to these explicit methods, the following implicit time integrators have also been implemented: implicit midpoint, and L-stable diagonally implicit Runge-Kutta methods of orders 2 through 4. The theoretical convergence rates of these methods have been computed and verified experimentally; regression tests exhibiting their capabilities have been added to the test suite. The second part of this task, implementation of the adaptive time stepper and Runge-Kutta error estimator, is under way and research into the optimal approaches is being conducted.

A new paradigm for implementing second-order, cell-centered finite volume (i.e., discontinuous Galerkin) type methods, based on piecewise linear gradient reconstruction and gradient limiting, has also been developed. This

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numerical method has made it possible to develop MOOSE codes that can accurately model convection-dominated flow and transport applications. A verification computational fluid dynamics application code using the rDG method was created and numerically verified against standard benchmark supersonic flow problems involving complex shock interactions, strong contact discontinuities, and rarefaction waves.

## Benefits to DOE

Each of these capabilities benefits existing programs in DOE in multiple ways. The CIVET application is a generic tool aimed at open and closed-source code development on multiple “git” hosting services. It was developed with stringent software quality assurance requirements in mind, such as those required by NQA-1. To the best of our knowledge, no other tool with these capabilities exists.

The Runge-Kutta implementation allows for more advanced analysis and comparison among several different numerical time integration schemes. While the Runge-Kutta implementations are not unique to MOOSE, the ability to switch among different methods and different orders with simple input file syntax makes advanced analysis, higher-fidelity simulation results, and parameter studies possible. Finally, the rDG prototype has significantly extended the application areas of MOOSE relevant to nuclear reactor/facility design and safety analysis in extreme conditions. This new paradigm has already been used on development of a multi-dimensional computational fluid dynamics code for analyzing the integrity of nuclear reactor containment vessels subjected to extreme thermal and overpressure-loading conditions. Future applications will include a fluid-solid interaction model involving coupled reactive fluid flow and solid structural mechanics in reactors. The development of a MOOSE fluid-solid interaction module is expected to provide unprecedented robustness and advantages over traditional fluid-solid interaction coupling strategies. All developments are being made available on a provisional basis to other research and development efforts using the MOOSE framework.

## References

- Tonks, M. R., D. Gaston, P. C. Millett, D. Andrs, and P. Talbot, 2012, “An object-oriented finite element framework for multiphysics phase field simulations,” *Computational Materials Science*, Vol. 51, No. 1, pp. 20–29.
- Williamson, R., J. Hales, S. Novascone, M. Tonks, D. Gaston, C. Permann, D. Andrs, and R. Martineau, 2012, “Multidimensional multiphysics simulation of nuclear fuel behavior,” *Journal of Nuclear Materials*, Vol. 423, No. 1, pp. 149–163.

## Publications

- Slaughter, A. E., J. W. Peterson, D. R. Gaston, C. J. Permann, D. Andrš, and J. M. Miller, 2015, “Continuous Integration for Concurrent MOOSE Framework and Application Development on GitHub,” *Journal of Open Research Software*, pp. 1–4, [Working towards Sustainable Software for Science: Practice and Experiences \(WSSSPE2\)](#).

## Presentations

- Xia, Y., 2016, “MOOSE solution methods for hyperbolic systems of partial differential equations,” presented at *MOOSE Training Workshop*, Idaho Falls, Idaho, May 2016 (INL/CON-16-38932.).

## Collaborations

National Laboratories: Argonne National Laboratory

## 16-058—Predicting Radiation-Induced Microstructural Change via Implementation and Validation of Multi-Scale Cluster Dynamics in MOOSE

Cody Permann,<sup>1</sup> Michael Short,<sup>1</sup> and Miaomiao Jin<sup>1</sup>

### General Project Description

The long time evolution of radiation-induced defects is a crucial issue for accurate prediction of material properties under irradiation. Studying the radiation defect cluster distribution can contribute to successful design of various radiation-resistant materials, not limited to nuclear applications, and predict their stability under irradiation. The most common tool for long-term evolution of radiation-induced microstructural change is cluster dynamics (CD), which tracks the flow of defects in and out of various defect clusters in size space. Calculations are carried out using rate theory construction, with both classical normal rate theory and production bias model taken into account for realism. Relevant parameters, including defect motilities and cluster absorption/emission rates, are derived mostly from atomic-level simulations. For quantities such as the binding energy of a vacancy to a void, for example, empirical thermodynamics-based correlations are used. However, most CD codes are not extendable to other situations, nor are they formulated in a way that allows for coupling of intermediate variables and final outputs to other multi-physics simulations. For example, if one were to accurately evolve the radiation-induced microstructure of silicon carbide using CD, it could be easily coupled to solid mechanics and thermal models of its degradation under irradiation. Finally, correct outputs of CD simulations are directly verifiable, using standard analytical techniques such as transmission electron microscopy, differential scanning calorimetry, and transient grating spectroscopy.

The governing equation for cluster dynamics is given below. This equation is adapted from rate theory describing the production, growth, and elimination of a defect cluster of several sizes with spatial resolution.

$$\frac{dC_j}{dt} = G_j + \sum \beta(j - k_1, k_1)C_{j-k_1}C_{k_1} - \sum \beta(j, k_2)C_jC_{k_2} + \sum \alpha(j, k_3)C_j - \sum \alpha(k_4, j)C_{k_4} + S_j$$

$C_j$  is the concentration of defect size  $j$ . On the right hand side, the first term  $G_j$  is the direct point defect or small cluster generation rate, the second to fourth terms represent absorption and emission, and the last term quantifies the elimination rate of defects at other microstructural sinks such as dislocations, grain boundaries, or free surfaces. Notice that  $G_j$  considers the clusters remaining after annealing radiation damage cascades. Defect clusters larger than 2 nm are visible under transmission electron microscopy for relatively easy verification. As a rough estimation, a 5-nm spherical cluster is approximately  $4 \times 10^4$ . It implies that to describe a general system of defect cluster distributions, at least tens of thousands of simultaneous, low-order size-space equations need to be solved at each spatial node.

### Summary

At this point, several parts of this project have been implemented in a prototype Multiphysics Object Oriented Simulation Environment (MOOSE)-based application, with some ahead of the proposed 3-year schedule:

1. Generation of defect clusters has been calculated based on binary collision approximation and annealing of damage cascades with the molecular dynamics code. The production rates of various cluster types and sizes have been pre-computed with spatial dependence.
2. A family of custom actions has been built into the MOOSE-based application to automatically generate constituent kernels for each variable. These constituent kernels each account for a single term from the master equation (given above) for each type and size of cluster on each spatial node. Flexible treatment of mobile and

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immobile defect clusters, production of defect clusters, and interaction coefficients have been completed. New parameters or radiation damage scenarios can be easily constructed by using the include/source files and corresponding input file. Absorption and emission rates for larger clusters are computed using well-established thermodynamic relations, while those for smaller clusters (which do not obey smooth functions) can be directly entered into the input file.

3. Technically, there is no upper limit to the size of the largest cluster. Nevertheless, the more defect species involved, the more difficult it becomes to solve the full set of equations numerically. Based on time scale and experimental observations, the largest size can be tailored to different cases directly in the input file. It is believed that more physics-based and accurate preconditioning of the problem will make the biggest impact toward increasing the computationally feasible maximum cluster size.
4. To decrease the computational burden of solving one equation for each defect species, one may group all sizes into a number of bins and solve one equation for each bin. A similar construction can be found in solving the neutron transport equation with the multi-group method. The basic idea is to use a normalizing function to calculate the weighted interaction coefficients for different groups and efficiently solve this new set of equations. Construction of this scheme has been finished and the normalizing function can be set to be any polynomial directly through the input file. However, the results are still in progress, because discrepancies exist in terms of different grouping schemes.

### Benefits to DOE

A full cluster dynamics capability benefits DOE by providing higher fidelity-scale bridging between traditional molecular dynamics simulations and mesoscale simulations. When complete, this bridging will enable a new class of simulations that is capable of furthering our understanding of various materials under irradiation, which directly benefit nuclear energy research. These capabilities will also further INL's lead nuclear laboratory leadership position in terms of modeling and simulations capabilities. When the final product is implemented in the MOOSE framework, it will be immediately available to other MOOSE-based applications; further increasing our simulation capabilities portfolio. Cluster dynamics implementation also challenges the upper limits on the sizes of our computational capabilities.

### Interns and Postdocs

Interns: Miaomiao Jin

Postdoc: Michael Short

### Collaborations

University: Massachusetts Institute of Technology

## 16-070—Characterization of Neutron Beamlines at NRAD

Aaron E. Craft,<sup>1</sup> Sam H. Giegel,<sup>2</sup> Glen C. Papaioannou,<sup>1</sup> Chad L. Pope,<sup>2</sup> and George R. Imel<sup>2</sup>

### General Project Description

The Neutron Radiography reactor (NRAD) sits beneath a large hot cell at the Hot Fuels Examination Facility and is designed specifically for neutron radiography of both highly radioactive and non-radioactive materials using two different neutron beams. The unique capabilities offered by NRAD and its neutron beams are well-suited for various research and development activities. However, information about these neutron beams is very limited. This project seeks to characterize the two neutron beams at the Hot Fuels Examination Facility's the East and North Radiography Stations to provide users with detailed information about these beams, including beam flux, spatial distribution, energy spectrum, divergence/collimation, and gamma content. This project will measure these characteristics for both the East and North Radiography Stations' neutron beams. The same beam characterization techniques will be extended to the neutron beam at the Transient Reactor Test Facility once operational. Characterization of these facilities will provide information that is essential for developing advanced digital neutron imaging capabilities, planning experiments and beamline upgrades, and providing data for validation of neutronics models.

### Summary

Characterization of neutron beams includes a suite of experimental measurements. The methods for performing these measurements are well-documented but can be technically challenging and quite involved. Users may require that the beam satisfy American Society for Testing and Materials standards as a high-quality neutron imaging facility. This analysis per American Society for Testing and Materials standards is relatively simple and is included in this project. Additionally, users may desire more detailed measures of beam quality such as beam uniformity, divergence, length-to-diameter ratio (L/D), neutron energy spectrum, and gamma content of the beam. This information is essential for extracting quantitative data derived from neutron radiographs generated at the NRAD facility. Some of these measurements require special devices and meticulous data processing and analysis.

This project is well-suited as a Masters of Science thesis topic, where it will receive the meticulous efforts of a full-time graduate student. Sam H. Giegel, a student from Idaho State University in the Nuclear Engineering Department, is the primary researcher of this project. Sam will be advised by professors at Idaho State University (Chad Pope and George Imel) and INL staff (Aaron Craft and Glen Papaioannou), who are experts in the facilities being characterized and the methods used. Sam has successfully completed his Bachelors of Science degree in Nuclear Engineering and is starting his coursework for his Masters of Science degree in the fall of 2016. As an intern during the summer of 2016, Sam worked alongside INL staff and became familiar with the people and facilities he will be working with. He has completed Radiation Worker II training, which will be required for some portions of this work. A literature review has been compiled of characterization techniques for neutron beams and previous characterization efforts. Materials required for the measurements have been identified and some materials have already been acquired. These activities during the first summer internship have prepared Sam and INL staff for carrying out the measurements for characterizing the neutron beams.

An array of gold foils will provide a total neutron flux profile and an array of cadmium-covered gold foils will provide a fast-spectrum neutron profile. A more detailed beam profile will be measured using transfer-method neutron radiography of the neutron beam, and the magnitude will be correlated from the foil activation measurements. The neutron energy spectrum will be measured using multi-foil activation and subsequent

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<sup>1</sup> Idaho National Laboratory

<sup>2</sup> Idaho State University

computational analysis and processing. An inventory has been compiled of activation foils already available at NRAD and additional materials for foil-activation measurements have been acquired. The beam flux, spatial distribution, and length-to-diameter ratio measurements will be repeated for the three length-to-diameter ratio settings at each of the two neutron beams. A gamma dose rate meter (ion chamber) has been acquired for measuring the gamma content of the neutron beams. The ion chamber has already been used to measure the gamma dose rate directly from irradiated fuel in the East Radiography Station. The ion chamber will be mounted at the image plane and a measurement will be taken with the reactor at full power.

After more than 20 years of inactivity, the North Radiography Station has been reactivated and is available for various neutron beam applications. This and another LDRD (i.e., 16-071) provided the impetus for reactivation of the North Radiography Station. The neutron beam profile of the North Radiography Station was measured during the summer of 2016 (ahead of schedule) using an array of gold foils and showed an average neutron flux of  $4.5 \times 10^6$  n/cm<sup>2</sup>s with good uniformity ( $2\sigma = 1.2 \times 10^5$  n/cm<sup>2</sup>s) over the  $7 \times 17$ -in. field of view. Future measurements will be taken over a larger  $14 \times 17$ -in. field of view.

The first year of the project focuses on preparing for the measurements that will be made during the second and third years. The project is slightly ahead of schedule because some measurements have already been accomplished during the first year. The project will be complete with the thesis project and publication in peer-reviewed journals that will serve as references for users and researchers.

### Benefits to DOE

Projects developing advanced nuclear fuels require neutron imaging capabilities to evaluate the condition of irradiated fuels and inform subsequent examinations. The neutron imaging capabilities provided by the neutron beams at NRAD directly support the primary mission of INL to advance nuclear energy in the United States. Characterization of these facilities will provide information that is essential for developing advanced digital neutron imaging capabilities, planning experiments and beamline upgrades, and providing data for validation of neutronics models. This project supports a graduate student, Sam Giegel, from Idaho State University.

### Interns and Postdocs

Intern: Sam H. Giegel

### Collaborations

University: Idaho State University



## 16-071—Evaluation of Advanced Digital Neutron Imaging Systems for PIE of Nuclear Fuel

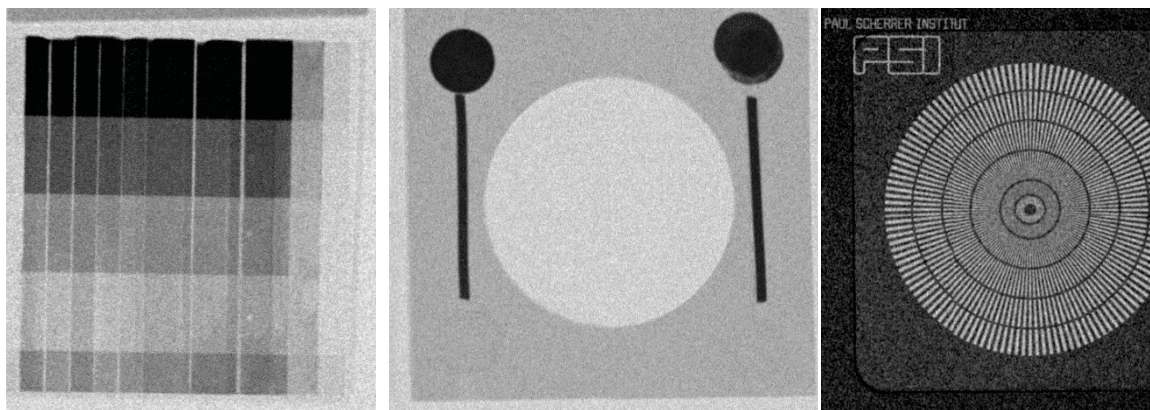
Aaron E. Craft,<sup>1</sup> Anton S. Tremsin,<sup>2</sup> and Glen C. Papaioannou<sup>1</sup>

### General Project Description

The current state-of-the-art for neutron imaging of irradiated fuel is the transfer method using converter foils with computed radiography plates or film, which is time-consuming and expensive. This project seeks to test advanced digital neutron imaging detectors for their applicability to evaluate irradiated nuclear fuel. The gamma sensitivity of digital neutron imaging systems has been the precluding factor for their use in this area. Micro-channel plate (MCP) detectors have low estimated gamma sensitivity that may allow their use for evaluating highly radioactive objects. For this project, a state-of-the-art system from the University of California-Berkeley has been tested at INL to evaluate the potential for using MCP-based imaging systems for evaluating irradiated nuclear fuel. If successful, an MCP system would be procured that would allow real-time digital neutron imaging and enable use of neutron tomography of irradiated fuel pins as a routine post-irradiation examination capability.

### Summary

This project seeks to test existing MCP systems for their ability to image irradiated objects by testing the detector response to gamma-contaminated neutron beams at the Neutron Radiography Reactor (NRAD) and to high gamma doses. Anton Tremsin develops state-of-the-art MCP systems that would be the best able of any MCP systems for imaging irradiated fuel. Thus, his detector was the primary focus for evaluation of these systems. MCP systems are already capable of imaging with neutrons; however, the impetus for these evaluations is to determine what level of gamma dose rate can the imaging system tolerate and still produce usable neutron images. Anton Tremsin visited INL to test his system. The detector was first installed in the North Radiography Station. Digital neutron images were acquired of image quality indicators (Figures 1). These images are the first fully digital neutron radiographs ever acquired at INL.



*Figure 1. Radiographs of an American Society of Testing and Materials sensitivity indicator (left) and beam purity indicator (right).*

This was the first time this system acquired neutron radiographs with a radial neutron beam with direct line-of-sight to the reactor core, which has significantly higher gamma radiation content compared to most

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<sup>2</sup> University of California-Berkeley

neutron-imaging beams. The gamma dose rate in the neutron beam was measured using an ion chamber and showed a dose rate of 100 R/hour at the image plane. The detector response was tested with lead filters of varying thickness (0 to 5 cm) positioned in the neutron beam to determine the effect on the neutron-to-gamma ratio. Irradiated fuel produces a gamma dose rate of approximately 1,000 to 10,000 R/hour, which would be reduced to less than 100 R/hour with some lead filtering (about 2.5 cm). The MCP system was able to produce neutron radiographs in this gamma field without significant degradation of image quality, lending optimism to the prospect of using the MCP system to image irradiated fuel.

After measurements at NRAD were complete, the system was transported to the Health Physics Instrumentation Laboratory and assembled in the Gamma Beam Laboratory to measure the detector response in gamma beam isotopic sources. The high dose rates from the isotopic sources (i.e., about 660 R/hour for  $^{137}\text{Cs}$  and about 130 R/hour for  $^{60}\text{Co}$ ) are similar to the dose rates expected from irradiated fuel, providing a means of gauging the potential for using an MCP detector system for imaging irradiated fuel. The detector response was tested with lead filters of varying thickness (i.e., 0 to 5 cm) positioned in the gamma beam.

Based on efforts during the first year of this project, it appears very plausible that an MCP neutron imaging system could be used to evaluate irradiated nuclear fuel, even in the high gamma fields from the NRAD neutron beams and the presence of irradiated nuclear fuel. Upcoming efforts seek to acquire digital neutron radiographs of an irradiated fuel pin using an MCP installed in NRAD's East Radiography Station at the Hot Fuel Examination Facility.

### Benefits to DOE

This project seeks to evaluate the applicability of advanced digital neutron imaging systems for post-irradiation examination of nuclear fuel. If successful, this project would lead to a system that would provide routine use of neutron computed tomography for post-irradiation examination of nuclear fuels and mark a significant improvement in the quality of data compared to current capabilities. This new capability would clearly demonstrate INL's leadership in nuclear fuels testing and promote the technical superiority of nuclear energy research and development in the United States.

### Collaborations

University: University of California, Berkeley

## 16-085—Production of Fluoroanion Targets for Accelerator Mass Spectrometry

Christopher Zarzana,<sup>1</sup> Gary Groenewold,<sup>1</sup> Michael Benson,<sup>1</sup> Kristyn Roscioli-Johnson,<sup>1</sup> Rika Hagiwara,<sup>2</sup> and Chien Wai<sup>3</sup>

### General Project Description

Detection of actinide element nuclides is critical for verifying compliance of nuclear facilities with international nonproliferation treaties. Current analysis methods require time-consuming and expensive sample preparation to achieve results acceptable to decision makers. Future analytical performance with markedly improved accuracy, precision, sensitivity, and faster sample throughput will be required to keep up with an increasingly complex nuclear proliferation environment. Accelerator mass spectrometry offers an order of magnitude increase in sensitivity for actinide analysis; however, current state-of-the-art still requires exacting separations that limit throughput. Additionally, actinides are currently analyzed as oxides; this introduces isobaric interference due to minor isotopes of oxygen, which limits the abundance sensitivity and precision. The objective of this project is to develop technology for rapid production of sample targets for actinide analysis using accelerator mass spectrometry through use of actinide fluoroanion salts produced using novel fluorinating ionic liquid and extracted using supercritical carbon dioxide. Actinide analysis using fluoroanion salts will increase sensitivity and precision through elimination of oxygen isobars. Additionally, manipulation of the degree of fluorination offers the potential for actinide separation in the ionization source, further reducing the need for chemical separation. The results of this project will be novel methods for rapid analysis of actinides using accelerator mass spectrometry, increasing the nation's capabilities in the nuclear nonproliferation sphere.



Figure 1.  $\text{UO}_2$  dissolved in EMImFHF.

### Summary

Uranium fluoroanions have been produced by dissolving  $\text{UO}_2$  in the fluorinating ionic liquid 1-ethyl-3-methylimidazolium fluorohydrogenate (EMImFHF) (Figure 1). The EMImFHF ionic liquid was provided through our university collaborator Professor Rika Hagiwara (Kyoto University). Fluoroanion production was successfully verified by electrospray-ionization mass spectrometry. Professor Hagiwara synthesized three more fluorinating ionic liquids: 1-butyl-3-methylimidazolium fluorohydrogenate, 1-butylpyridinium fluorohydrogenate, and 1-butyl-1-methylpyrrolidinium fluorohydrogenate. All three ionic liquids dissolve  $\text{UO}_2$  but to significantly different degrees, suggesting the structure of the ionic liquid cation plays a large role in dissolution chemistry.

To examine the influence of cation on dissolution, we traveled to the FELIX laboratory in Nijmegen, Netherlands, to perform gas-phase infrared (IR) multi-photon dissociation (IRMPD) measurements on

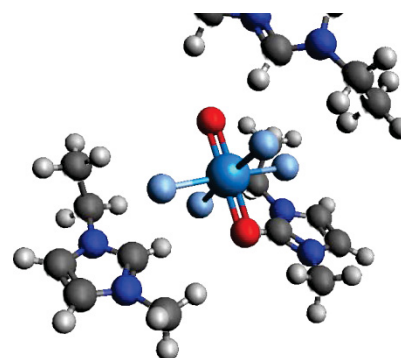


Figure 2. Gas-phase structure of  $\text{UO}_2\text{F}_4^{2-}$  stabilized by three EMIm<sup>+</sup> cations. The structure was calculated using density functional theory and matches well with experimental IRMPD data.

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<sup>2</sup> Kyoto University, Japan

<sup>3</sup> University of Idaho

clusters containing the EMIm cation and several fluoroanions, including  $\text{UF}_6^-$ ,  $\text{UO}_2\text{F}_3^-$ ,  $\text{UO}_2\text{F}_4^{2-}$ ,  $\text{ZrF}_5^-$ , and  $\text{ZrF}_6^{2-}$  (Figure 2). These studies revealed that the anions and cations bind in specific ways depending on structure, which suggests the fluorination chemistry could be controlled by varying structure; this is one of the main hypotheses of this project. We will continue to test this hypothesis with the three newly synthesized ionic liquids and anticipate returning to the FELIX laboratory to conduct more gas-phase IRMPD measurements. Additionally, these results have suggested further experiments that could provide new insight into the mesoscopic ordering of ionic liquids (an area that is challenging to study) and open it to further exploration. We also made the first measurements of the IR absorption frequencies of several fundamental vibrational modes of  $\text{UF}_6^-$ ,  $\text{UF}_5^-$ ,  $\text{UO}_2\text{F}_3^-$ , and  $\text{ZrF}_5^-$  as part of our experimental campaign at the FELIX laboratory. We are currently comparing these measurements to computational chemistry theory with the aim to enhance the accuracy of the calculations.

In addition to direct scientific accomplishments, we have conducted materials suitability tests that verify the chemical compatibility of the ionic liquids we are studying with the super-critical carbon dioxide systems we plan on using in FY 2017 and FY 2018. We have also begun preparation of work control documents to enable measurement of actinide-containing samples with an ion-trap mass spectrometer located at the Central Facilities Area. This instrument will support future investigations of neptunium and plutonium in these ionic liquid systems.

## Benefits to DOE

This project is developing novel methods for rapid actinide analysis that are urgently needed by DOE and other national security customers. Rapid, inexpensive actinide analysis methods will greatly enhance the ability of the international community to ensure compliance with international treaties, increasing global security. Additionally, this project is also exploring the fundamental actinide chemistry needed to develop these methods, furthering DOE's basic science mission.

## Publications

Zarzana, C. A., G. S. Groenewold, M. Benson, K. Johnson-Roscioli, J. Martens, J. Oomens, R. Hagiwara, and T. Tsuda, 2016, "Spectroscopic and Mass Spectral Characterization of Uranium Fluoroanions Formed from an Ionic Liquid," *Plutonium Futures – The Science 2016*, Baden Baden, Germany, September 18 through 22, 2016.

## Collaborations

University: University of Idaho and Kyoto University

## 16-096—Supporting Operator Performance and Situation Awareness in Highly Automated Nuclear Power Plants

Katya Le Blanc<sup>1</sup> and Johanna Oxstrand<sup>1</sup>

### General Project Description

The objective of this project is to investigate the impact of high levels of automation in complex, safety critical systems and to demonstrate ways to enable optimal situation awareness for human operators in those systems. Specifically, this research will demonstrate how to design human-automation interaction to support optimal situation awareness, workload, and plant performance in a multi-unit small modular reactor.

### Summary

In FY 2016, INL researchers worked with NuScale personnel to design realistic scenarios for investigating operator performance under a variety of levels of automation. NuScale power has defined a preliminary human automation interaction design, where many of the plant's functions are fully automated, but some functions have an intermediate level of automation. INL researchers developed three interfaces for a boron dilution task with varying levels of automated support for the operator. The research worked with operations, engineering, and human factors staff at NuScale to design an experimental test for evaluating the effect of each of the three interfaces on human performance and situational awareness. One of the interfaces developed by the INL team has been adopted by NuScale as a template for all automation processes in the control room. This interface will be updated based on the results of this and future studies, resulting in a design that optimally supports operators in automated tasks.

INL researchers supported methodology development for the workload analysis NuScale conducted as part of their request for exemption to the Nuclear Regulatory Commission (NRC) staffing rule. We advised NuScale on how to perform and analyze several common workload measurement tools and provided several suggestions on how to develop criteria for characterization of acceptable workload. NuScale adopted several of our suggestions, including using workload at an existing operating plant as baseline criteria for workload comparison. NuScale noted that the discussions we had on workload methodology helped them develop their own expertise and enhanced their ability to successfully present their approach to NRC. The workload analysis is an important part of NuScale's request for exemption to the NRC staffing rule, which is one of the greatest challenges to licensing the NuScale design. Supporting the workload analysis directly supports NuScale's licensing efforts.

The researchers designed an experiment to test the dilution display's effect on performance, situational awareness, and workload. The researchers worked closely with NuScale operations engineers, human factors engineers, and simulator engineers to design scenarios and performance measures for the experiment. Testing of the experimental protocol and initial data collection began the week of June 13, 2016, and continued through September 20, 2016. Experimental data collection was slightly delayed due to challenges with the NuScale simulator and delays in their workload analysis for NRC that took priority over this work. Data collection will be complete by December 2016.

### Benefits to DOE

- The results of this work support design of human automation interaction in the nuclear industry and will also inform use of advanced technologies that may be installed in existing light water reactors as part of control room upgrades and design of systems installed to support grid modernization and hybrid energy systems.

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<sup>1</sup> Idaho National Laboratory



- This work comprises scientifically rigorous research in close collaboration with industry. We meet several key challenges in conducting scientific research in full-scale simulator studies, thus advancing the science of human performance measurement in the nuclear context.
- This work supports licensing of a small modular reactor and demonstrates an effective collaboration between INL and NuScale that benefits the entire nuclear industry and any industry that has human-automation interaction challenges:
  - NuScale provides the platform, facilities, operational expertise, and operators for conducting high-quality research to understand human automation interaction
  - Interfaces developed and tested in this work will be implemented in the NuScale control room and the concepts can be applied to related work in other Office of Nuclear Energy programs
  - Support of NuScale's licensing directly supports the DOE mission of deploying a small modular reactor.

## **Publications**

The paper titled, "Measuring Workload: Consequences of Weightings when Combining NASA TLX Subscales," is in final preparation and will be submitted to Human Factors in September 2016.

The researchers will prepare a paper summarizing the experimental results and will submit it to Safety Science in December 2016.

## ***Presentations***

Presented project to 2016 Nuclear Science and Technology Advisory committee on May 17, 2016.

## **Interns and Postdocs**

Intern: Rachael Hill

Postdoc: Wei Zhang

## **Collaborations**

University: University of Idaho

Industry: NuScale



## 16-098—Nuclear Nonproliferation Applications of $^{14}\text{C}$ Analyses by Accelerator Mass Spectrometry

Mathew Snow,<sup>1</sup> John Olson,<sup>1</sup> Mary Adamic,<sup>1</sup> Matthew Watrous,<sup>1</sup> and John Southon<sup>2</sup>

### General Project Description

In January 2015, INL installed a 0.5-MV accelerator mass spectrometer (AMS). The INL AMS is ideally suited for ultra-trace isotopic analyses supporting national security applications, including nuclear forensics and nonproliferation. This project seeks to develop INL's sample preparation and AMS analysis capability to analyze the isotopic composition of carbon containing samples for ultratrace  $^{14}\text{C}$ .

### Summary

FY 2016 efforts were dedicated to two general categories: (1) INL staff training at University of California-Irvine and construction of an INL  $^{14}\text{C}$  sample preparation manifold system, and (2) preliminary method development and validation of INL's sample preparation capabilities for simple organic samples.

#### **INL Staff Training and $^{14}\text{C}$ Sample Preparation Manifold Construction:**

During FY 2016, INL scientists received training in current state-of-the-art  $^{14}\text{C}$  AMS sample preparation techniques under the direction of the University California-Irvine scientists Dr. John Southon and Dr. Xaime Xu (leading experts in  $^{14}\text{C}$  AMS analyses). Following this training, the INL team developed and constructed a gas phase  $^{14}\text{C}$  sample preparation manifold system (Figure 1). Construction of this system was required in order to stand up the ultra-trace  $^{14}\text{C}$  AMS analysis capability at INL. The current manifold configuration has a combustion line for conversion of organic samples to carbon dioxide and a graphitization line for cryogenic purification of carbon dioxide prior to sample graphitization and  $^{14}\text{C}$  AMS analysis.



*Figure 1. INL's  $^{14}\text{C}$  sample preparation manifold system constructed under this project.*

<sup>1</sup> Idaho National Laboratory

<sup>2</sup> University of California-Irvine

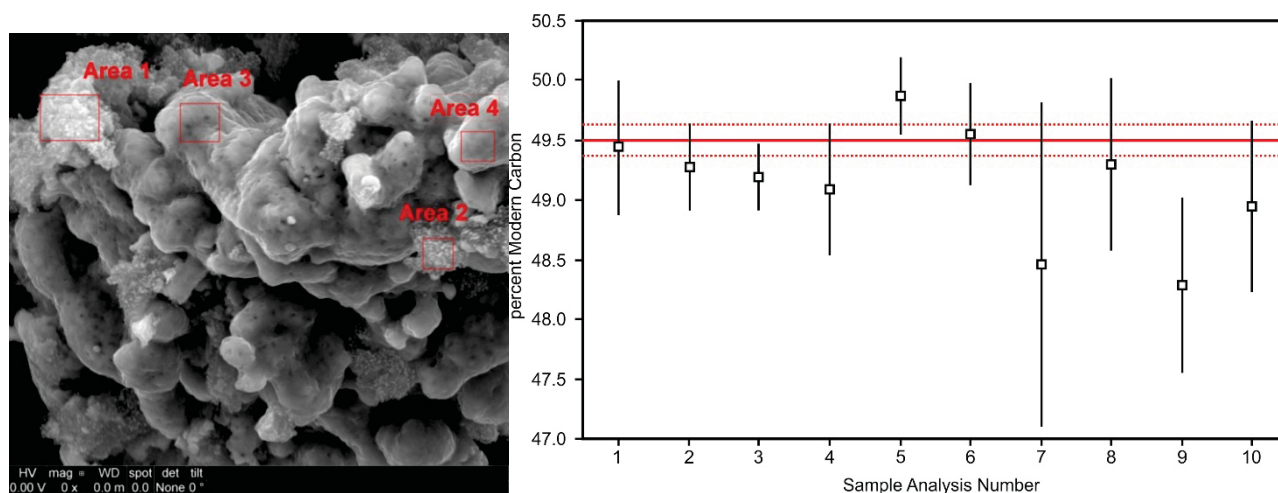


Figure 2. (left) Scanning electron microscope micrographs showing the formation of purified carbon species on the surface of an iron catalyst. (right) Preliminary  $^{14}\text{C}$  analysis data for the standard International Atomic Energy Agency-C7 (oxalic acid). Red solid and dashed lines represent the certificate value and associated uncertainty. INL measurements show production of high yields of purified carbon with the correct  $^{14}\text{C}$  content.

### Preliminary Evaluation of Analysis Capabilities for Organic Samples

Preliminary method development and validations for analyses of simple organic samples were performed in FY 2016 using International Atomic Energy Agency and National Institute of Standards of Technology oxalic acid standards, with the current optimized method results shown in Figure 2. INL measurements for International Atomic Energy Agency-C7 are in agreement with the certificate value for this material, demonstrating initial success in standing up the ultra-trace  $^{14}\text{C}$  AMS sample preparation and analysis capability at INL. Further refinements in the measurement precision and extension of current analysis capabilities to include more complex matrices (including complex environmental samples, inorganic samples, and air samples) will continue throughout the lifecycle of this project.

### Benefits to DOE

$^{14}\text{C}$  sample preparation/analysis capability development at INL enables application of this isotope to support a variety of National and Homeland Security missions, including nuclear forensics and nonproliferation. Additional potential future applications of this capability range from long-term environmental stewardship supporting advanced nuclear fuel cycles, clean alternative energy sources, and evaluations of global climate change.

### Publications

Snow, M. S., 2016, "Isotopic Signature and Standards Development Supporting Nuclear Forensics and Nonproliferation," *National Technical Nuclear Forensics Academic/National Laboratory Collaboration Meeting*, Los Alamos, New Mexico, August 3, 2016.

### Collaborations

University: University of California, Irvine

## **16-129—Application of Radioactive Isotope Dilution Technique for Measurement of Molten Salt Mass for the Electrochemical Recycling Process**

Shelly Li<sup>1</sup> and Jeff Sanders<sup>1</sup>

### **General Project Description**

Measuring the total mass of high-temperature molten salt in containers is a critical step for the International Atomic Energy Agency in its safeguards program for electrochemical recycling plants and molten salt reactors through nuclear material accountancy. A much needed measurement technology for determining molten salt mass is yet to be developed. During FY 2016, this project sought to investigate application of a radioactive isotopic dilution technique to determine molten salt mass in containers of irregular shapes. The technical goal was to develop a technology for near real nuclear material accountancy with good measurement accuracy that can be used to enhance the safeguards of electrochemical recycling plants and molten salt reactors.

### **Summary**

During FY 2016, all proposed research activities have been completed. A patent application is in drafting and, until that patent application is filed, it is important to protect the detailed research results from public disclosure.

Research activities were conducted in collaboration with Ohio State University. The experimental work and radioactivity measurements were performed at the Ohio State University Nuclear Reactor Laboratory. The feasibility study results are promising.

### **Benefits to DOE**

If the proposed research is successful, it will be a breakthrough in safeguards measurements for electrochemical recycling processes and will significantly improve the state-of-the-art. The proposed technique may also be used for other safeguards purposes and applications that are of interest to DOE (e.g., for molten salt reactor systems).

### **Publications**

#### ***Presentations***

Poster presentation at National and Homeland Security Strategic Advisory Committee meeting, June 30, 2016, a non-public forum.

### **Invention Disclosures, Patents, Copyrights**

Invention Disclosure Record (IDR) BA-887, “Application of Radioactive Isotope Dilution on Determination of Molten Salt Mass for Electrochemical Recycling Process,” filed on January 4, 2016.

INL Technology Deployment has elected title from DOE to the IDR BA-887 for a patent application on May 3, 2016. DOE Case #: S-143,824. The patent application is in drafting.

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<sup>1</sup> Idaho National Laboratory

## 16-149—In-Core Qualification of Developmental Instrumentation

Joshua Daw<sup>1</sup> and Joe Palmer<sup>1</sup>

### General Project Description

The purpose of this project is to perform ex-core characterization of several sensors under development for in-core use. This ex-core testing is necessary in anticipation of potential in-core tests, because it will give a basis for comparison with in-core results. Primarily, the sensors to be tested are temperature sensors. Several of these sensors are based on physical principles that can be used for measuring many parameters in addition to temperature (i.e. ultrasound and fiber optics). Therefore, successful demonstration of these sensors will lead to further sensor development for an extended range of parameters.

### Summary

Progress to this point has been primarily limited to fabrication and testing of prototype ultrasonic thermometers (UTs) (see Figure1).

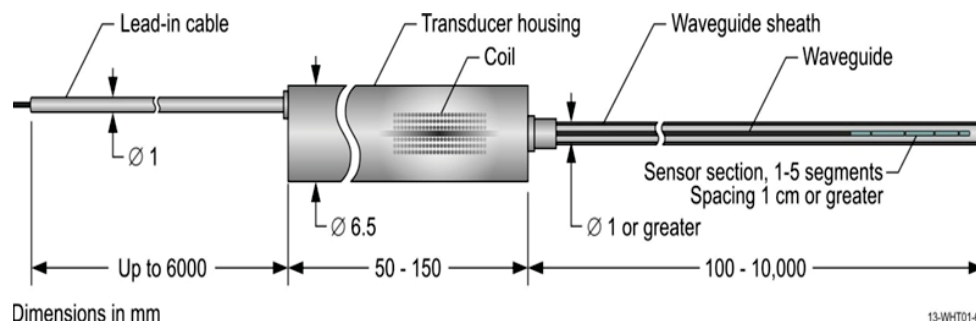


Figure 1. Schematic of UT.

Several variations of the UT have been fabricated or are in the process of fabrication. The configurations to be evaluated are described in Table 1.

Table 1. UT design variations.

UT Prototype Designation	Sensor Material	No/ of Sensor Segments	Segment Length	Status
Inc 606	Inconel 606	1	8 cm	Initial testing complete
CP Ti	Commercially pure titanium	1	8 cm	Initial testing complete
304 SS	304 stainless steel	1	8 cm	Initial testing complete
Mo-1	Molybdenum	1	8 cm	Initial testing complete
Mo-2	Molybdenum	1	4 cm	Fabrication complete
Mo-3	Molybdenum	1	2 cm	Fabrication complete
Mo-4	Molybdenum	1	1 cm	Fabrication complete
Mo-5	Molybdenum	2	1 cm	Fabrication started
Mo-6	Molybdenum	4	1 cm	Fabrication started

<sup>1</sup> Idaho National Laboratory

UT Prototype Designation	Sensor Material	No/ of Sensor Segments	Segment Length	Status
Mo-7	Molybdenum	8	1 cm	Fabrication started

Testing to-date has been limited to general performance testing. Development of calibration curves and characterization of temperature accuracy and resolution has not been possible due to extremely late delivery of some necessary testing equipment. This equipment has arrived and is currently being set up and debugged. Testing of UTs will continue through FY 2016.

Fiber optic temperature sensors, based on extrinsic Fabry-Perot interferometry, have been provided by Luna Innovations. The specific sensors provided by Luna are coated in a polyimide, which limits their operating temperatures to about 300°C. The sensors may be tested to about 800°C, but this will volatilize the coating and reduce the durability of the sensors. This test will be performed and will be considered destructive to the tested sensor. A Luna-developed optical backscatter reflectometer (OBR) has been leased in order to test the sensors. The OBR is the signal conditioner for optical fiber sensors. Luna believes that the OBR, not the fibers themselves, will enable their sensors to be used in a radiation environment. A Luna engineer delivered the sensors and OBR to INL and instructed INL researchers in use of the system. This testing will begin in September 2016 and be completed by October 30, 2016. Luna has also offered to supply a 1-m long fiber to test continuously distributed measurement of temperature (also using the leased OBR).

The acoustic gas monitor to be provided by French Alternative Energies and Atomic Energy Commission (CEA) will not be available for testing. CEA feels that the sensor has been well qualified for in-reactor use, but it currently does not have the ability to implement recommended upgrades to the sensor design. As a replacement for testing, CEA has provided an intrinsic distributed fiber optic temperature sensor of their design, based on Fiber-Bragg gratings and the hardware required to operate the sensor and record data. The Fiber Bragg gratings sensor has 10 evenly spaced gratings along a 10-cm sensor length, allowing measurement of a temperature gradient. Maximum test temperature for this sensor is about 800°C.

The National Priorities List-developed Johnson noise thermometer is not yet ready for testing. It is anticipated that testing will be carried out in FY 2017 pending completion of National Priorities List development.

The National Priorities List-developed Pt/Pd thermocouple was initially of interest based on the belief that irradiation resistance would be high due to the transmutation behavior of Pd. More thorough investigation has revealed that this is likely not the case, because the transmutation chain of Pd is more complicated than initially described. The TC is still of interest for lower fluence irradiation experiments, as a drift resistant reference thermocouple for calibrating other instruments, and may be tested in FY 2017.

Several issues have been encountered during the course of this project. Issues with equipment and delayed deliveries have caused delays to fabrication and testing of UTs. A more serious concern is with the ability of the collaborating organizations to provide the sensors they have offered. CEA was unable to provide the acoustic gas monitor and instead has provided a fiber optic sensor. This change was discussed with the LDRD committee and was determined to be acceptable.

## Benefits to DOE

Development of advanced in-core instrumentation is critical to DOE's energy security mission to provide world-class facilities for advancing nuclear science and technology. Successful completion of this effort will result in ex-core characterization of several new sensors that could be deployed during irradiation testing in U.S. high-flux materials test reactors, such as ATR, and the Transient Test Reactor facility when it is restarted.

## **Interns and Postdocs**

Postdocs: Richard Skifton and Pattrick Calderoni

## **Collaborations**

University: Massachusetts Institute of Technology

Industry: National Physical Laboratory and LUNA Innovations

National Laboratories: The French Alternative Energies and Atomic Energy Commission



## 16-187—Micro-Scale Technique to Evaluate Grain Boundary Cohesion of Irradiated Alloys

Chao Jiang,<sup>1</sup> Wen Jiang,<sup>1</sup> Brian Jaques,<sup>1</sup> Indrajit Charit,<sup>2</sup> and Ray Fertig<sup>1</sup>

### General Project Description

Metallic alloys are widely used or planned for use as structural and cladding materials in current and future reactors. Under irradiation, grain boundary (GB) cohesion strength decreases due to interaction with defects and impurities, leading to intergranular fracture and embrittlement of alloys. The objective of this project is to develop a technique for quantifying GB cohesion and its impact on fracture behavior in irradiated alloys, by utilizing transmission electron microscopic (TEM) in situ cantilever testing in concert with multi-scale modeling. The TEM in situ cantilever testing is a novel approach for studying the real-time mechanical response of materials. It will be used in this work for studying intergranular fracture behavior in several irradiated iron-based ferritic alloys and providing key information to link atomistic-level events with mesoscale/macroscopic mechanical properties. The Multi-Physics Object-Oriented Simulation Environment (MOOSE)-based cohesive zone model (CZM) and extended finite element method (XFEM) for intergranular fracture of irradiated ferritic alloys will be developed in this work by utilizing atomistic results as inputs and experimental results for validation.

### Summary

During the first year of this project (FY 2016), the focus of the experimental efforts was on developing capabilities for measuring GB cohesion strength using the microscopy and characterization suite, specifically the focused ion beam and in situ PicoIndenter in TEM, at the Center for Advanced Energy Studies. By modifying previous micro-mechanical methods used to compress micro-pillars, micro-cantilever beams were fabricated (as shown in Figure 1) with the expectation to isolate GB cohesion strengths. The notched cantilever beams were produced using a focused ion beam to create a tensile fracture. Cantilever beams were fabricated and tested from a model Fe-<sup>9</sup>Cr oxide dispersion-strengthened (ODS) alloy. Of these cantilever beams, 15 were tested from bulk material and six were tested from ODS irradiated to 3 dpa at 500°C. In addition to the cantilever setup, we are also considering different designs (such as four-point bending) to facilitate fracture observation. We will also try a cube-corner tip instead of a flat punch tip in order to limit out-of-plane bending.

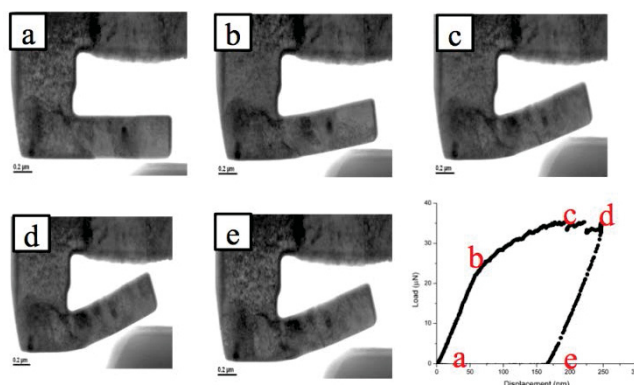


Figure 1. Fe-<sup>9</sup>Cr ODS micro-cantilever bend test conducted at the Center for Advanced Energy Studies.

<sup>1</sup> Idaho National Laboratory

<sup>2</sup> University of Idaho

Two alloys, Fe-<sup>9</sup>Cr and Fe-<sup>20</sup>Cr-<sup>5</sup>Al-<sup>0.5</sup>Ti-<sup>0.5</sup>Y<sub>2</sub>O<sub>3</sub> (similar to MA 956 composition), are currently being synthesized from their constituent powders using high-energy ball milling that is followed by spark plasma sintering. All spark plasma sintering runs are carried out at 80-MPa pressure and a heating/cooling rate of 100°C/min. X-ray diffraction of select batches of ball-milled powder are being conducted in order to know the degree of alloying, crystalline size, lattice parameters, and lattice strain. TEM experiments of selected spark plasma sintered specimens are planned to study the microstructural characteristics, including grain size, GBs, second phases (e.g., size and distribution) and so forth.

On the atomistic scale, molecular dynamics simulations have been conducted to calculate the cohesive strength of clean GBs in bcc Fe, defined as the minimum energy to fracture a GB. A few interatomic potentials for molecular dynamics modeling of Fe-based alloys have been identified and assessment of their suitability for our modeling is in progress. From the results shown in Table 1, it can be concluded that GBs with lower energies are also stronger against decohesion. The molecular dynamics-calculated GB cohesive strength values are directly usable in CZM and XFEM models.

*Table 1. Cohesive strengths and energies of symmetric tilt Fe GBs from molecular dynamics simulations.*

GB type	$\Sigma 3<111>$	$\Sigma 5<100>$	$\Sigma 5<100>$	$\Sigma 9<110>$
Cohesive Strength (J/m <sup>2</sup> )	3.51	2.55	2.72	2.60
GB Energy (J/m <sup>2</sup> )	0.27	1.10	1.00	1.32

On the continuum scale, initial code development in the MOOSE framework has been conducted. To integrate CZM with XFEM, a new Elem-Elem constraint system has been implemented in the MOOSE framework. This new constraint allows users to incorporate general cohesive laws at crack surfaces. For CZM development, a novel tri-linear cohesive law has been developed to improve numerical convergence without adding artificial viscous regularization terms. This law gives correct energy dissipation and has been demonstrated good agreement with experiment results. To further consider irradiation effects, the evolution of a cohesive zone with irradiation damage will be developed and implemented in MOOSE.

## Benefits to DOE

The radiation-induced degradation of material mechanical properties poses serious limitations to nuclear energy applications. The capabilities of TEM in situ mechanical testing and MOOSE-based fracture models developed for this work will help understand and predict the performance of materials in reactors. In turn, it would enable safer and more economical nuclear energy in the future. Results from this project will contribute to the DOE's leading role on research of materials behavior and performance in radiation environments.

## Publications

Yano, K. H. and J. P. Wharry, 2016, "TEM In Situ Cantilever Testing to Assess Grain Cohesion in Irradiated ODS," *Materials Science and Technology 2016*, Salt Lake City, Utah, October 2016.

## Interns and Postdocs

Postdoc: Wen Jiang

## Collaborations

University: Boise State University, University of Idaho, and University of Wyoming

## 16P6-003FP—Phenomena Identification and Ranking Table Technique Applied to the MEGA-POWER Heat Pipe Reactor Concept

James Sterbentz,<sup>1</sup> James Werner,<sup>1</sup> Michael McKellar,<sup>1</sup> Andrew Hummel,<sup>1</sup> John Kennedy,<sup>1</sup> John Biersdorf,<sup>1</sup> and Richard Wright<sup>1</sup>

### General Project Description

In recent years, the U.S. Department of Defense (DOD) has become increasingly interested in the potential of small nuclear reactors for military use. DOD's interest in small reactors stems mainly from two critical vulnerabilities it has identified in its infrastructure and operations: (1) the dependence of U.S. military bases on the fragile civilian electrical grid and (2) the challenge of safely and reliably supplying energy to troops in forward operating locations.

A small (i.e., 1 to 10-MWe) heat-pipe-cooled, fast-spectrum nuclear reactor has been identified as a candidate for forward operating applications and other possible applications. The point design serves as a basis for this project and was provided by a group of scientists from Los Alamos National Laboratory. This group of experts has done a significant amount of high-quality work to characterize the reactor, but has not yet performed an in-depth safety analysis of the concept.

The Los Alamos National Laboratory reactor (i.e., the MEGA-POWER reactor) operates at 5 MW (thermal), with a power conversion system capable of generating up to 2 MWe for 5 years. The reactor consists of a stainless steel (SS316) monolith structure containing 5.22 metric tons of 19.75% enriched uranium-oxide ( $\text{UO}_2$ ) fuel pellets and with liquid metal potassium heat pipes operating at 677°C. The heat pipes remove the heat from the block as the liquid in the heat pipe is vaporized; no pumps or valves are required. The heat is subsequently deposited in the condenser region of the heat pipe. The condenser region is sized to accommodate multiple heat exchangers, such as one for power conversion and two for redundant decay heat removal. The reactor uses an alumina ( $\text{Al}_2\text{O}_3$ ) neutron reflector, with 12 embedded control drums that contain an arc of boron-carbide poison on the drum. The active part of the core is about 1 m flat-to-flat and 1.5 m high. The outer diameter of the  $\text{Al}_2\text{O}_3$  reflector is 1.5 m. In the proposed concept, the monolith core is fabricated in six identical segments (see Figure 1).

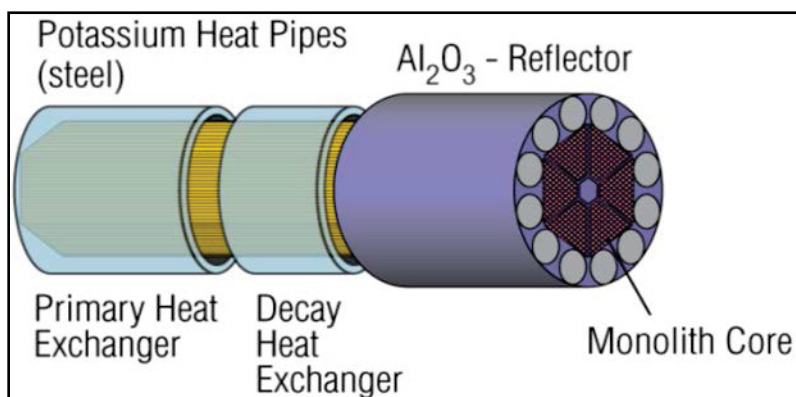


Figure 1. MEGA-POWER heat pipe reactor concept schematic.

<sup>1</sup> Idaho National Laboratory

### Summary

The purpose of this project is to identify areas of potential concern that will lead to experiments and analyses directed toward “a proof of principle” or early determination of the utility of this new reactor concept that would have a direct positive impact on utilization of nuclear energy and our national defense.

This project has been executed from the outset and follows three parallel efforts. The first is ongoing development of our own independent assessment of the reactor concept through development of analytical thermal and neutronic models to allow the team to be in a position to assess the design and make recommendations regarding robustness and development paths. The second is development of a dynamic operating model of the power conversion system unit and supporting equipment for the MEGA-POWER reactor concept. A power conversion design using the ASPEN HYSYS process modeling software has been developed. This analysis provides the team information regarding how much power can be produced and any feedback characteristics that may affect reactor operation as a result of planned or unplanned transients. So far, redesign efforts have upped the electrical output from 782 kW to 1.58 MW. The third effort is developing and applying the Phenomena Identification and Ranking Table (PIRT) technique to the MEGA-POWER concept. The team is identifying phenomena that dominate specific safety-related issues. This approach of applying the PIRT methodology was chosen to give the INL team an overall systems view of the concept. The nine-step PIRT process is being used to develop the major topical areas for which the PIRT process will be applied. Four PIRTs are being developed: (1) reactor normal and accident conditions, (2) heat pipe normal and accident operations, (3) reactor materials and fabrication issues, and (4) power conversion system.

### Benefits to DOE

The core mission at INL is to discover, demonstrate, and secure innovative nuclear energy solutions, clean energy options, and critical infrastructure. The MEGA-POWER reactor concept could be an important technology that would enhance DOD and homeland security aspects by enabling the rapid prototyping and development of a first-of-kind small, low-power reactor that would enhance security and provide an essential energy source to forward operating bases or strategically important national facilities. Innovative reactor designs (such as the MEGA-POWER reactor) advance INL’s nuclear computational and experimental capabilities. INL’s vision is to change the world’s energy future and secure our critical infrastructure and, in doing so, contribute to the nation’s economic opportunity. This project advances all of these desired outcomes as the U.S. national lead nuclear energy laboratory. This project aligns many of INL’s broad range of research capabilities with the goal of providing world leadership in nuclear research, development, demonstration, and deployment.





## Enabling Clean Energy Deployment

- 14-078—Extended Stability Gamma-Gamma Prime Containing Nickel-Base Alloys
- 14-079—Second Generation Switchable Polarity Solvent Draw Solutes for Forward Osmosis
- 14-086—Development of a Microgrid/Smartgrid Testbed for ESL and Super Lab Initiative with Load Variability Characterization and Control for Renewable Energy Integration
- 14-095—In Situ Measurement of Electrolyte Chemistry in Battery Cells During Operation
- 14-106—Understanding the Growth of Ultra-Long Carbon Nanotubes
- 15-039—Transient Modeling of Integrated Nuclear Energy Systems with Thermal Energy Storage and Component Aging and Preliminary Model Validation via Experiment
- 15-125—Phosphoranimines for Advanced Battery Applications
- 15-128—Microstructural Evolution and Mesoscale Coupled Flow-Reaction-Fracturing Processes in Organic Rich Nanoporous Shales
- 15-135—Dynamic Simulations for Large-Scale Electric Power Networks in Real-Time Environment using Multiple RTDS
- 15-140—Expanding the Utility of Advanced Chemical Physics Models for Electrolytes
- 15-146—Tailoring the Kinetic Function of a Surface through the Electronic Effects of Nanoscale Architecture
- 16-002—Advanced Carbon Feedstock Processing Using Ionic Liquids
- 16-176—Development of Direct Carbon Fuel Cells
- 16-215—Electrochemical Manufacturing Processes
- 16P6-002FP—Kinetic-Based Scale-Up Science for an Energy Efficient Route to Ethylene



## 14-078—Extended Stability Gamma-Gamma Prime Containing Nickel-Base Alloys

S. Meher,<sup>1</sup> L. K. Aagesen,<sup>1</sup> T. M. Pollock,<sup>2</sup> L. J. Carroll,<sup>1</sup> and M. C. Carroll<sup>1</sup>

### General Project Description

Increased efficiencies for electricity and power generation through concentrating solar, nuclear, and fossil energy continue to demand improved efficiencies. These increasing efficiencies require materials that exceed the properties of the available state-of-the-art. Fortunately, material requirements for specific hot section components are similar: an extended stability alloy capable of operating for long times and, at times, reaching temperatures of up to 1000°C. The objective of this work is to develop microstructural strategies for  $\gamma$ - $\gamma'$  superalloys through targeted chemistry and processing to enable extended stability.

### Summary

Three classes of superalloys (i.e., Ni-base, NiCr-base, and CoNi-base) were investigated. Detailed microstructural characterization of all three classes of alloys at nanometer scales followed the high-temperature exposures. Critical experimental inputs were integrated and utilized with a computational kinetic modeling tool to simulate the microstructure of the alloys subjected to high-temperature exposures. Characterization of the phase compositions and precipitate volume fraction, size, and shape after aging was completed using transmission electron microscopy, scanning electron microscopy, and atom probe tomography. Microstructural modeling was carried out by phase field modeling and CALPHAD-based software.

### *Hierarchical $\gamma$ - $\gamma'$ Microstructures*

An alternative pathway for enhanced coarsening resistance of ordered  $\gamma'$  precipitates, which is important for extended stability, is to form homogeneously distributed nano-scale disordered  $\gamma$  precipitates. This microstructural evolution has been observed in one of the investigated alloys at 800°C. Atom probe tomography results (Figure 1) have confirmed supersaturation with respect to cobalt and ruthenium in the  $\gamma'$  phase as the driving force for nucleation of  $\gamma$  precipitates. The sluggish coarsening kinetics of  $\gamma$  precipitates due to presence of refractory elements such as tantalum, tungsten, and rhenium has a pertinent hierarchical effect on the possible sluggish coarsening rate of  $\gamma'$  precipitates. Additionally, phase field modeling (Figure 1) supports the coarsening behaviors of  $\gamma$  precipitates in both the dendrite and interdendritic region and was used to identify factors that affect the stability of the  $\gamma$  precipitates within the  $\gamma'$  phase.

### *Low $\gamma$ - $\gamma'$ Interfacial Energy*

A second pathway to enhanced coarsening resistance is through precipitate distributions with low interfacial energy. Isothermal annealing at 900°C for times up to 256 hours has been studied experimentally and through simulation for two experimental multicomponent  $\gamma$ - $\gamma'$  nickel-base alloys with spherical precipitate morphologies. The coarsening behavior of the  $\gamma'$  precipitates is in accordance with the matrix diffusion-limited coarsening, with coarsening rate constants similar to those reported for other superalloys. By exploiting a modified Lifshitz-Slyozov-Wagner relationship between the coarsening rate constant and the  $\gamma$ - $\gamma'$  interfacial energy, the interfacial energies are estimated from experimentally determined phase compositions and measured precipitate coarsening. The estimates of the interfacial energies, validated through kinetic simulations, are much lower than typically reported for other multicomponent  $\gamma$ - $\gamma'$  nickel alloys.

<sup>1</sup> Idaho National Laboratory

<sup>2</sup> University of California Santa Barbara

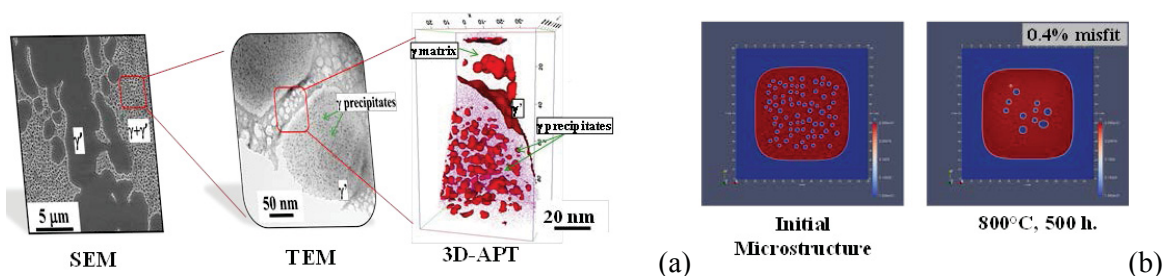


Figure 1. (a) A collage of images from different microscopic techniques showing nano-scale hierarchical precipitation and (b) phase-field modeling simulation results showing the evolution of secondary  $\gamma'$  precipitates in the dendritic region.

### Alloying Chemistry

Alloying strategies for extended microstructural stability of multi-component  $\gamma$ - $\gamma'$  CoNi-base alloys, based on control of solute partitioning between ordered  $\gamma'$  precipitates and the disordered  $\gamma$  matrix, have also been investigated as a means of achieved extended stability. The unique shift in solute partitioning in these alloys, when compared to that in simpler Co-base alloys, derived from changes in the site substitution of solutes as the relative amounts of cobalt and nickel change, highlight new opportunities for development of tailored alloys.

### Benefits to DOE

This project has developed  $\gamma$ - $\gamma'$  superalloys chemistry and microstructural design strategies, promising for high-temperature, extended-service durations to introduce inherent long-term stability by introducing new phase transformation pathways. The alternate pathways for achieving coarsening resistance for  $\gamma$ - $\gamma'$  superalloys investigated as part of this project provide fundamental strategies for future development of high-temperature materials for energy applications.

### Publications

- Meher, S., L. J. Carroll, T. M. Pollock, and M. C. Carroll, 2016, "Solute partitioning in multi-component  $\gamma/\gamma'$  Co-Ni-base superalloys with near-zero lattice misfit," *Scripta Materialia*, Vol. 113, pp. 185-189.
- Meher, S., L. K. Aagasen, L. J. Carroll, M.C Carroll, and T. M. Pollock, 2016, "Understanding of Inverse Coarsening of  $\gamma'$  precipitates in Ni-base Superalloys," *Microscopy and Microanalysis*, Vol. 22, No. S3, pp. 1258-1259.
- Meher, S., M. C. Carroll, T. M. Pollock, and L. J. Carroll, "Precipitate coarsening in  $\gamma$ - $\gamma'$  nickel-base superalloys with low interphase interfacial energies," manuscript in preparation.
- Meher, S., L. K. Aagasen, T. M. Pollock, M. C. Carroll, and L. J. Carroll, "Co-existence of  $\gamma$  and  $\gamma'$  precipitates in Ru and Re containing nickel-base superalloys," manuscript in preparation.
- Meher, S., L. K. Aagasen, M. C. Carroll, T. M. Pollock, and L. J. Carroll, "Coarsening of bimodal  $\gamma'$  distribution in high refractory containing nickel base superalloys," manuscript in preparation.

### Presentations

- "Correlative orientation microscopy and atom probe tomography on nickel and cobalt-base superalloys," *Microscopy & Microanalysis 2016 Conference*, Columbus, Ohio, invited.
- "Inverse Coarsening of  $\gamma'$  precipitates in Nickel-base Superalloys," *TMS 2016 Annual Conference*, Nashville, Tennessee.

“Extended microstructural stability in high refractory containing nickel-base superalloys,” *TMS 2015 Annual Conference*, Orlando, Florida.

### **Interns and Postdocs**

Postdoc: Subhashish Meher

### **Collaborations**

University: University of California Santa Barbara

## 14-079— Second Generation Switchable Polarity Solvent Draw Solutes for Forward Osmosis

Aaron D. Wilson,<sup>1</sup> Christopher J. Orme,<sup>1</sup> Josh McNally,<sup>1</sup> Frederick F. Stewart,<sup>1</sup> and Jeffrey R. McCutcheon<sup>2</sup>

### General Project Description

The switchable polarity solvent (SPS) forward osmosis (FO) process can purify water from extremely concentrated feeds containing components such as salts, organics, inorganics, and biologics. INL has demonstrated that a dimethylcyclohexylamine SPS draw solution can provide osmotic flux against a 226,000 ppm tds NaCl solution. SPS FO functions at a fraction (i.e., less than 0.5) of the cost of existing methods through a thermolytic cycle that avoids distillation of water but still uses heat energy, with heat costing a fraction of the electrical energy used in reverse osmosis. The drawback of this initial demonstration of SPS FO is the aggressive solvent characteristics of dimethylcyclohexylamine, which damage membranes and a wide range of rubbers and plastics. Advancement of SPS FO to industrial implementation requires an SPS that retains the high osmotic pressures of dimethylcyclohexylamine without the material incompatibilities. In this project, materials will be developed as draw solutes for FO systems that have high osmotic pressures, low viscosities, and reasonable costs. These materials will require properties similar to previously studied materials, while avoiding material degradation, reduced toxicity, reduced membrane permeation, and improved SPS switching behavior. This work will include synthesis and characterization of amines. With a viable SPS, an integrated demonstration of an SPS FO process will be developed based on the most effective SPS candidate.

### Summary

The first effort in this project was to complete and publish a structure-function model of SPS's capability to form concentrated solutions (Wilson and Stewart 2014). This structure function model allowed us to identify our Gen II SPS class based on 1-cyclohexylpiperidine. 1-cyclohexylpiperidine has a higher molecular weight than the SPS used to demonstrate the SPS FO process. The higher molecular weight provides a number of advantages, including (a) reduced vapor pressure/odor; (b) better kinetics, lower temperatures, and more complete polar to non-polar phase transitions (degassing); (c) lower water solubility in the non-polar form; (d) better materials compatibility; and (e) lower likelihood of passing through membranes for FO/reverse osmosis applications. The Gen II SPS adds all of these advantages while retaining the high osmotic pressure and low production cost of N,N-dimethylcyclohexylamines. Battelle Energy Alliance, LLC, was granted a patent for the method of use of Gen II SPS in FO water treatment (US 9,399,194). Our group also published a paper highlighting the draw solute performance metrics of 1-cyclohexylpiperidine (Orme and Wilson 2015). This work was followed up by a computational chemistry density functional theory study by Dr. Joshua McNally to identify and understand the intramolecular structures that influenced intermolecular and inter-ion interactions that produce bulk properties and behaviors (McNally and Wilson 2015). Dr. Joshua McNally has since been converted to full-time staff; this is due, in part, to his work on this project.

We also conducted a concentration dependent study of Gen II and two control SPSs. This work has resulted in a published paper addressing mass transport properties and solution-state speciation that is generalized to all SPSs (Wilson and Orme 2015). These concentration-dependent properties are fundamental for processing design and optimization of any application of SPS.

We have also generated ( $\pm$ )-trans-N,N,N',N'-tertamethyl-1,2-Diaminocyclohexane at the 300-g scale and demonstrated that they are effective SPS materials. The diamine can reach 80 wt% in its polar form before

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forming a crystalline material matching the performance of the best monoamines. During the next year, concentration performance metrics will be obtained for the diamine and compared to our Gen II SPS.

Our collaboration with Professor McCutcheon has yielded two significant results. The first was analysis of the fundamental energy associated with pressure-retarded osmosis and removing/concentrating TDS from/in a solution (Reimund et al. 2015). The second was a membrane/SPS compatibility that addressed water flux data, reverse solute flux, and solute/membrane parameters for several membranes (Reimund et al. 2016). A third paper exploring membrane performance with related draw solutions via the pressure-retarded osmosis process is anticipated in the near future (Wilson 2016).

We previously obtained an x-ray crystal structure of a N,N-dimethylcyclohexylammonium bicarbonate. This structure featured a hydrogen-bonded bicarbonate dimer. Recently, Frank Weinhold at the University of Wisconsin and Roger Klein proposed the existence of anti-electrostatic hydrogen-bond complexes that would conclusively demonstrate a dominant covalent component of the hydrogen bond. Our bicarbonate dimer is an example of an anti-electrostatic hydrogen bond and, based on a variety of experimental evidence, appears to be present in the solution state. This would be the first example of an anti-electrostatic hydrogen bond complex found in aqueous solution. Joshua McNally traveled to Oak Ridge National Laboratory's Spallation Neutron Source User Facility to obtain a neutron structure of our bicarbonate dimer. The results should influence our understanding of the hydrogen bond, which will have a wide impact on our understanding of chemistry from the classroom to the research laboratory.

Based on our published work (including papers from this project), we were contacted by a startup, Rebound Technology, who is working in the field of modest to large-scale refrigeration. The Rebound interaction has developed into a collaboration that resulted in a funded Phase I STTR project. This collaboration is built around a new field of use for SPS that would not have been possible without the fundamental work developed in this project.

In addition to the conversion of Joshua McNally to a full-time employee, there have been two recent hires relevant to developing and expanding INL staff. Postdoc, Dr. Birendra Adhikari, chemical engineer, CU Boulder 2015, and undergraduate Catherine Hrbac, chemical engineer, MIT 2015, were hired through this project to expand our expertise in chemical engineering. Their chemical engineering skills are required for developing fundamentals processes associated with SPS FO and other water treatment technologies.

## Benefits to DOE

Water treatment and desalination is an energy intensive process with room for improvement. Reducing this energy cost would meet DOE's mission; however, the connection between water and energy runs deeper. The current energy infrastructure requires water to produce energy (e.g., evaporative cooling and biomass and algae production) and obtain energy resources (e.g., well injection [fracking] water). Water used by energy industries eventually requires treatment (e.g., oil and gas water, blowdown water, and many others). Mission impact beyond DOE would be best described in terms of energy, water, and food security. The technology under development may also have application for water production at forward-operating bases. This project is expected to develop scalable water treatment technology that can reduce the cost of producing and processing waters relevant to the energy industry and military.

SPSs are a rapidly developing field of research. Our current work allows us to advance their application to water treatment and other fields of use, as well as publish fundamental work on SPSs that have implications on very fundamental concepts, including electrolyte theory and the hydrogen bond. This work allows us to meet DOE science and applied missions.

## References

- McNally, J. S. and A. D. Wilson, 2015, “Density Functional Theory Analysis of Steric Impacts on Switchable Polarity Solvent (SPS),” *Journal of Chemical Physics*, Vol. 119, pp. 6766-6775, DOI: 10.1021/acs.jpcc.5b03167.
- Orme, C. J. and A. D. Wilson, 2015, “1-Cyclohexylpiperidine as a Thermolytic Draw Solute for Engineered Osmosis,” *Desalination*, Vol. 371, pp. 126-133, DOI: 10.1016/j.desal.2015.05.024.
- Reimund, K. K., J. R. McCutcheon, F. F. Stewart, and A. D. Wilson, 2015, “Thermodynamic analysis of energy density in pressure retarded osmosis: The impact of solution volumes and costs,” *Journal of Membrane Science*, Vol. 487, pp. 240-248, DOI: 10.1016/j.memsci.2015.03.076.
- Reimund, K. K., B. J. Coscia, J. T. Arena, A. D. Wilson, and J. R. McCutcheon, 2016, “Characterization and Membrane Stability Study for the Switchable Polarity Solvent N,N-Dimethylcyclohexylamine as a Draw Solute in Forward Osmosis,” *Journal of Membrane Science*, Vol. 501, pp. 93-99, DOI:10.1016/j.memsci.2015.10.039.
- Wilson, A. D., 2016, “Design of the next-generation FO draw solution,” Chapter 6 in *Rational Design of Next-Generation Nanomaterials and Nanodevices for Water Applications*, pp. 103–130, IWA Publishing, August 2016, ISBN-10: 1780406851, ISBN-13: 978-1780406855.
- Wilson, A. D. and F. F. Stewart, 2014, “Structure-Function Study of Tertiary Amines as Switchable Polarity Solvents,” *RCS Advances* 2014, Vol. 4, pp. 11039-11049.
- Wilson, A. D., and C. J. Orme, 2015, “Concentration Dependent Speciation and Mass Transport Properties of Switchable Polarity Solvents,” *RCS Advances*, Vol. 5, pp. 7740-7751, DOI: 10.1039/C4RA08558B.

## Publications

- McNally, J. S. and A. D. Wilson, 2015, “Density Functional Theory Analysis of Steric Impacts on Switchable Polarity Solvent (SPS),” *Journal of Chemical Physics B*, Vol. 119, pp. 6766-6775, DOI: 10.1021/acs.jpcc.5b03167.
- Orme, C. J. and A. D. Wilson, 2015, “1-Cyclohexylpiperidine as a Thermolytic Draw Solute for Engineered Osmosis,” *Desalination*, Vol. 371, pp. 126-133, DOI: 10.1016/j.desal.2015.05.024.
- Reimund, K. K., J. R. McCutcheon, F. F. Stewart, and A. D. Wilson, 2015, “Thermodynamic analysis of energy density in pressure retarded osmosis: The impact of solution volumes and costs,” *Journal of Membrane Science*, Vol. 487, pp. 240-248, DOI: 10.1016/j.memsci.2015.03.076.
- Reimund, K. K., B. J. Coscia, J. T. Arena, A. D. Wilson, and J. R. McCutcheon, 2016, “Characterization and Membrane Stability Study for the Switchable Polarity Solvent N,N-Dimethylcyclohexylamine as a Draw Solute in Forward Osmosis,” *Journal of Membrane Science*, Vol. 501, pp. 93-99, DOI:10.1016/j.memsci.2015.10.039.
- Wilson, A. D. and F. F. Stewart, 2014, “Structure-Function Study of Tertiary Amines as Switchable Polarity Solvents,” *RCS Advances* 2014, Vol. 4, pp. 11039-11049.
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- Wilson, A. D., 2016, “Design of the next-generation FO draw solution,” Chapter 6 in *Rational Design of Next-Generation Nanomaterials and Nanodevices for Water Applications*, pp. 103–130, IWA Publishing, August 2016, ISBN-10: 1780406851, ISBN-13: 978-1780406855.



## Presentations

- Orme, McNally, Wendt, and Wilson, 2015, “1-Cyclohexylpiperidine-CO<sub>2</sub> Switchable Polarity Solvent (SPS) Draw Solution Performance Results,” *North American Membrane Society (NAMS) 25<sup>th</sup> Annual Meeting*, Boston, Massachusetts, June 2015.
- Reimund, Coscia, Wilson, and McCutcheon, 2014, “Forward Osmosis Flux Performance and Membrane Compatibility Studies of the Dimethylcyclohexyl Ammonium Hydrogen Carbonate Switchable Polarity Solvent (SPS) Draw Solute,” *North American Membrane Society (NAMS) 24<sup>th</sup> Annual Meeting*, Houston, Texas, June 2014.
- Wendt, Mines, Orme, and Wilson, 2015, “Development of Switchable Polarity Solvent Draw Solutes,” *International Forward Osmosis Association (IFOA) World Summit*, Vancouver, Canada, September 2015, invited.
- Wilson, 2014, “Working at a National Laboratory,” *North American Membrane Society (NAMS) 24th Annual Meeting*, Houston, Texas, June 2014, invited speaker.
- Wilson and Orme, 2014, “Further Studies of Switchable Polarity Solvent (SPS) As Draw Solutes for Forward Osmosis,” *North American Membrane Society (NAMS) 24<sup>th</sup> Annual Meeting*, Houston, Texas, June 2014.
- Wilson, 2015, “Design and Function of Switchable Polarity Solvent (SPS) Draw Solutes,” *INL Early Career Research Symposium*, July 2015.
- Wilson, 2015, “SPS FO Water Treatment Technology,” a web based “Water ReUse” Innovation Showcase put on Emerging Tech Accelerator sponsored by National Institute of Standards and Technology’s manufacturing extension partnership. SPS FO technology was one of two “next generation water treatment technologies” that were presented to the 266 attendees representing 199 unique commercial entities, July 2015, invited.
- Wilson, 2015, “Advancing New Research in Water Innovation for the Future at INL,” *Advancing Water Efficiency in Downstream Refinery, Petrochemical, and Chemical Processing*, Sugarland, Texas, November 2015 (invited speaker and discussion panelist).
- Wilson, Orme, Jones, Hrbac, Adhikari, Wendt, and Mines, 2015, “Switchable Polarity Solvent Forward Osmosis Process Development,” *2015 AIChE Annual Meeting, INL Advanced Manufacturing Group’s Energy-Water Nexus Workshop*, Salt Lake City, Utah, November 2015, invited speaker.
- Wilson, 2016, “Forward Osmosis Water Technology Demonstration,” *DOE National Laboratory Environmental Stewardship Day* on Capitol Hill, Washington D.C., September 2016.
- Wilson, Hagood, and Austin, 2016, “Forward Osmosis Water Technology Demonstration,” *White House Water Summit*, Eisenhower Executive Office Building, Washington D.C., March 22, 2016.

## Invention Disclosures, Patents, Copyrights

- US 9,399,194, “Draw Solutions and Related Methods for Treating a Liquid,” 2016, Aaron Wilson and Christopher Orme (BA-808 2396).

## Interns and Postdocs

Interns: Kevin K. Reimund

Postdocs: Birendra Adhikari and Joshua S. McNally

**Collaborations**

University: University of Connecticut

## 14-086—Development of a Microgrid/Smartgrid Testbed for ESL and Super Lab Initiative with Load Variability Characterization and Control for Renewable Energy Integration

Kurt Myers,<sup>1</sup> Jason Bush,<sup>1</sup> Jake Gentle,<sup>1</sup> Bob Turk,<sup>1</sup> Porter Hill,<sup>1</sup> Manish Mohanpurkar,<sup>1</sup> and Rob Hovsopian<sup>1</sup>

### General Project Description

With an increasing demand for renewable and distributed generation integration, the requirements for legacy grid and market structures are rapidly exceeding current capabilities. The ability to understand, test, and model the system dynamics of real-world microgrids and distributed energy resources (DER) at multiple scales and geographic locations is essential to successfully increasing renewable penetration. The developing micro/smartgrid platform at INL addresses these issues through development of DER grid and microgrid testing, controls research and development (R&D), and modeling capabilities in an integrated, collaborative environment. To achieve these goals, INL has been developing and implementing an open-source smartgrid platform with integrated control systems and component interaction R&D, developing scalable grid models from acquired data; integrating and testing high penetration levels of inverter-based renewable energy generation, battery storage, and load control; and working with industry and government to begin hardware and software integration testing for future microgrid and DER applications. Additionally, this platform allows direct testing and demonstration of various topologies of hardware (including smart inverters and other power electronics systems); electrical, mechanical, and controls interactions; and grid energy storage use-case testing, which may be difficult or impossible to accurately simulate when fully coupled to a campus microgrid or full utility grid system.

This project research determines the contribution of variability from renewable generation and various load types and how management, regulation, and control of grid systems can benefit through optimized integration, improved use-case control algorithms, and testing of new and existing load, storage, and generation resource technologies. It provides DOE with a state-of-the-art testbed for analyzing and dissecting the dynamic, coupled behavior of renewable generation, energy storage, traditional generation, and load. The research into controllers and architecture for energy and storage management schemes (that provide user/grid reliability, security/resiliency, and enhanced grid and user economics) is developing state-of-the-art capabilities and intellectual property that help identify competitive-cost resource applications within well-designed electricity markets or microgrids.

### Summary

Due to direct funding receipt in early FY 2016, this project was closed. The microgrid testbed and R&D developed during this project will be leveraged for direct project use for various Department of Defense microgrid projects and other R&D work, including integration of new flow battery systems and testing of energy storage and power electronics/controls systems for other industry, utility, and DOE research efforts. From a follow-on work perspective, this project has been very successful in building DOE capabilities and bringing in new R&D work and work potential. Some project accomplishments for the R&D duration include the following:

1. Developed multiple control and management algorithms, architecture, and power systems models for grids with high penetration levels of variable generation and energy storage systems (ESSs) for both large grid and microgrid applications.
2. Demonstrated stable primary and secondary control layers and architecture for systems with at least 50 to 75% average renewable energy content.

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3. Integrated, developed, and conducted use-case measurement and experimentation with secondary use and other ESS systems and developed diagnostic/prognostic findings and needs for continuing R&D. Battery efficiencies, use-case performance applicability, remaining cycle life potential, and system economics baselines were characterized.
4. Multiple microgrid electrical, controls, and economic models were developed in software modeling systems, including Homer, EasyPower, LabView, RTDS, and RS/PSCAD, for use in steady-state and dynamic systems networks and larger system simulations. These models have enabled further study of DER system interactions, system economics and cost targets modeling, and hardware/software linkages with other laboratory assets and external partners.

### Benefits to DOE

This research supports the DOE goal of solving key technological challenges in making clean energy efficient and affordable, while maintaining reliability and resiliency. This project was beneficial to DOE in developing smartgrid/microgrid and DER integration technologies and testing and modeling assets that will assist others in meeting federal goals for clean energy implementation, integration, and grid modernization. DOE is pursuing multiple efforts in DER, energy storage, grid modernization, and renewable energy integration; this project has positioned INL to provide research and demonstration avenues across these areas in conjunction with linkages to utilities, industry, and Department of Defense partnerships and efforts. This work is also benefiting the Department of Defense, utility, industry, and other partners through test and research input into their planning, development, business/economic dispatch, and operational models and processes.

### Publications

- Bhattarai, B., K. Myers, et al., 2015, “Overvoltage mitigation using coordinated control of demand response and grid-tied rooftop photovoltaics,” *3rd IEEE SusTech2015*.
- Bhattarai, B., K. Myers, et al., 2016, “Active control of thermostatic loads for economic and technical support to distribution grids,” *IEEE PES General Meeting*.
- Bhattarai, B., K. Myers, and J. Bush, 2016, “Reducing Demand Charges and Onsite Generation Variability Using Behind-the-Meter Energy Storage,” *IEEE SusTech2016*.

### Presentations

- Hill, P., K. Myers, B. Turk, and J. Bush, 2015, “Metering for Power Systems and Microgrid Planning,” and “Use of Wind Energy in INL and Other Microgrid Systems,” *AWEA Windpower 2015*, poster presentations.
- Myers, K., 2015, “Renewable Energy/Power Systems Development and Integration: Testing and Use-Case Experiences for Grid-Tied/Micro/Island Grid Applications of Battery Energy Storage,” *The Battery Show 2015 conference*.
- Myers, K., 2015, “Renewable Energy Development and Integration, Microgrids, ESS, and Power Systems,” *NWPPA Power Supply conference*, August 2015.

### Collaborations

University: Boise State University, Florida State University, and Idaho State University

Industry: Idaho Power Company, ON Semiconductor, and POWER Engineers

## 14-095—In Situ Measurement of Electrolyte Chemistry in Battery Cells During Operation

Gary S. Groenewold,<sup>1</sup> David Jamison,<sup>1</sup> Cathy Rae,<sup>1</sup> Kristyn Johnson,<sup>1</sup> Chris A. Zarzana,<sup>1</sup> and Kevin L. Gering<sup>1</sup>

### General Project Description

Lithium (Li) ion batteries are remarkable power storage devices capable of delivering power over repeated discharge-recharge cycles and a range of environmental conditions. Nevertheless, Li ion battery performance is eventually degraded as a result of chemical and physical changes within the cells. If time and use-dependent behavior were known, it is likely that informed approaches to mitigate deterioration and improve performance would emerge. Unfortunately, battery cells are sealed to prevent reaction with the ambient atmosphere; therefore, it is difficult to measure quantitative chemical changes occurring within the cell. The objective of this research is to develop Li ion battery cells that will enable chemical interrogation in the form of sub-microliter sampling and analysis and in situ vibrational spectroscopy, without compromising the integrity or function of the cell as it undergoes discharge/recharge cycling. It is hypothesized that diagnostic changes in the chemical composition of the electrolyte mixtures can be measured as the battery ages and progresses toward eventual failure. Achieving this objective is challenging because the total quantity of electrolyte present in a typical cell is small. However, the project leverages recent development of mass spectrometry analysis strategies that require only a small fraction of a microliter; a salient challenge is to manufacture a cell with ports that support periodic sample collection.

### Summary

Li ion battery cells were designed and fabricated with a sampling valve that enabled acquisition of either vapors or sub-microliter samples. The cells displayed reproducible discharge/recharge cycling, from  $V_{\max} = 4.7$  to  $V_{\min} = 2.7$ ; 15 cells of this type were fabricated. Experiments were conducted where operating temperature and electrolyte composition were varied and enabled collection of samples. In the majority of cells, failure was achieved after a few dozen cycling events, which enabled examination of the corresponding electrolyte.

Two different gas chromatography/mass spectrometry methodologies were developed for measuring electrolyte concentrations. Sub-microliter samples were analyzed by direct injection after dilution by greater than five orders of magnitude and head space was collected using a solid phase micro-extraction device to sample the residual vapor in the micro-sampling valve. This enabled measurement of the concentrations of the carbonate electrolytes and impurities generated by electrolyte degradation, which suggested mechanistic pathways, specifically ester hydrolysis.

Extensive research at INL has resulted in development of fire retardant additives that have the potential to improve the safety of the Li ion batteries without compromising their performance. However, measurement of these compounds is challenging, particularly for coin cells, where the overall volume is exceptionally small. Coin cells were fabricated that contained fire retardant additive compounds and that were designed to be harvested to enable collection of microscopic samples (i.e., less than 100 nanoliters). Ordinarily, this volume would create problems in handling and analysis; however, to overcome these limitations, the samples were injected into an electrospray ionization mass spectrometer using a technique called induction-based fluidics. A droplet of 35 nanoliters was extruded from a syringe with a non-conducting, fused silica capillary tip and situated directly in front of the spectrometer aperture. High potential was then applied to the capillary, which induced charge on the surfaces of both the droplet and the capillary. Charge repulsion caused the droplet to be launched from the surface and into the aperture of the mass spectrometer (Figure 1). The fast scanning spectrometer enabled collection of

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mass spectra that showed distribution of molecules in the fire retardant additive to the electrolyte, which was found to contain a number of congeners that varied in the extent of fluorination. Because the fire retardant originated from the electrolyte, significant Li was present in the droplet sample and appeared complexed to the fire retardant molecules. The tendency to bind Li was found to be correlated with the extent of fluorination of the base molecule and the more fluorinated Li complexes also displayed a stronger tendency to form extended structures where the carbonate electrolytes were also complexed. It is hypothesized that this may prove to be a sensitive indicator of additive function.

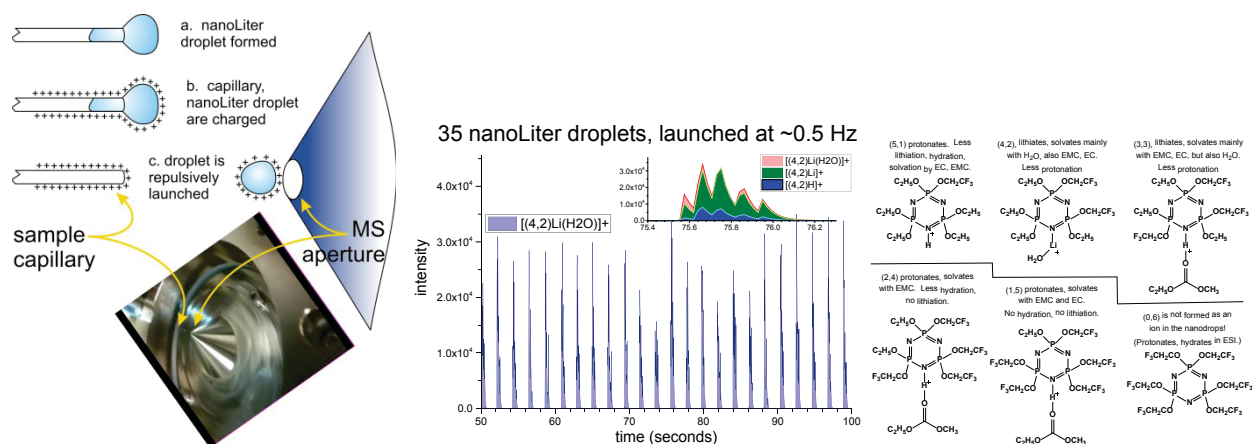


Figure 1. (left) Schematic depiction of a nanoliter droplet launched into a mass spectrometer aperture; (center) temporal profile of mass spectral response to repeated nanoliters, and (right) variable complexation chemistry dependent on the extent of fluorination.

## Benefits to DOE

The project holds excellent potential for providing a much-improved understanding of electrolyte chemistry that impacts battery performance and ultimate lifetime. The range of DOE and government functions that would be benefitted by improved battery performance would be vast and include applications in national security, energy storage, and environmental remediation actions. Consequent improvements in battery technology would have wide-reaching effects that benefit virtually all government agencies.

## Publications

### Presentations

Sauter III, A. D., D. Sauter, K. Johnson, C. Zarzana, G. S. Groenewold, R. Ross, and P. Limbach, 2016, "Droplets for IBF Mass Spectroscopy and Analytical Chemistry," 64<sup>th</sup> American Society for Mass Spectrometry Conference, San Antonio, Texas, June 5 through 9, 2016.

## Interns and Postdocs

Intern: Bethany Kursten



## 14-106—Understanding the Growth of Ultra-Long Carbon Nanotubes

Joshua J. Kane<sup>1</sup>

### General Project Description

Carbon nanotubes (CNT) have extraordinary properties, including a tensile strength 100 times greater than steel, a thermal conductivity rivaled only by the purest of diamonds, and an electrical conductivity similar to copper but capable of carrying significantly higher current densities. These properties make it a desirable material for future composites, coatings and films, energy storage, and environmental applications.

Current CNT applications are not able to take full advantage of these unique properties due to the inability to grow aligned arrays of macroscopic-length CNTs. A majority of industrial applications currently use a random dispersion of CNTs within a bulk material to slightly augment the material properties of the composite. The major factor limiting CNT growth is poisoning and deactivation of the catalyst used. The purpose of this project is to understand some of the experimental factors affecting deactivation kinetics at a continuum level.

### Summary

#### **FY 2014**

Project achievements in FY 2014 included design, acquisition, and receipt of an experimental system capable of measuring the rate of carbon accumulation within a chemical vapor deposition chamber.

#### **FY 2015**

During FY 2015, significant strides were made toward enabling investigation of the mechanism of CNT growth. The first major milestone in FY 2015 was assembly and optimization of the custom-built CVD reactor for in situ measurements of gas composition. Gas analysis used a quadrupole mass spectrometer. In situ analysis during CNT growth experiments is rare in CNT literature and sorely needed according to some of the leading experts in CNT growth kinetics.<sup>1</sup> In situ analysis of gas composition during growth experiments will provide additional insight to various aspects of CNT growth and deactivation that typically are unavailable in “common/traditional” growth experiments. The second major milestone for FY 2015 was development of a standard, reproducible method for growth of ultra-long CNT to begin probing at major variables affecting CNT growth and deactivation.

#### **FY 2016**

Experimental investigations focused on the effects of inlet gas composition on CNT growth. Within this investigation, two different effects were studied in more detail. The first study concentrated on understanding the effect of homogenous gas phase reactions on the growth of CNTs. Figure 1a shows three CNT arrays grown under identical conditions but different flow rates (300 sccm, 900 sccm, and 2,700 sccm top to bottom, respectively). Figures 1b through 1d show the typical structure observed at various magnifications within the CNT array. It was found that changing the flow rate changed the gas phase composition over the CNT growth substrate. Increasing the flow rate decreased the dwell time of a gas within the reactor prior to passing over the CNT catalyst. This effectively decreased the time available for the inlet  $H_2(g)$  and  $C_2H_4(g)$  to react and form  $CH_4(g)$ . At the temperatures investigated,  $CH_4(g)$  cracks over the catalyst substrate surface and deposits significant amounts of amorphous carbon on the catalyst. Over time, a buildup of amorphous carbon could lead to deactivation of the catalyst. There are no known studies on the effects of gas phase reactions on CNT growth in the vast CNT publication literature. The formation of  $CH_4(g)$  from  $H_2(g)$  and  $C_2H_4(g)$  could explain many of the inconsistencies

<sup>1</sup> Idaho National Laboratory

in growth throughout the literature for various researchers using identical experimental conditions in different experimental reactors. A publication on the experimental results of this study is anticipated in early FY 2017.

Adding small amounts of  $\text{H}_2\text{O}(\text{g})$  to the inlet gas stream has been demonstrated (in the CNT literature) to prolong CNT growth. Literature suggests this is a result of  $\text{H}_2\text{O}(\text{g})$  reacting with amorphous carbon on the catalyst surface, effectively keeping metal active sites available for further CNT growth. However, moisture does not prolong CNT growth indefinitely. The second experimental study investigated the effects of  $\text{H}_2\text{O}(\text{g})$  on CNT growth. Experimental work from this study suggests that  $\text{H}_2\text{O}(\text{g})$  over time changes the oxidation state of the catalyst, forming a metal oxide that is unable to sustain CNT growth.

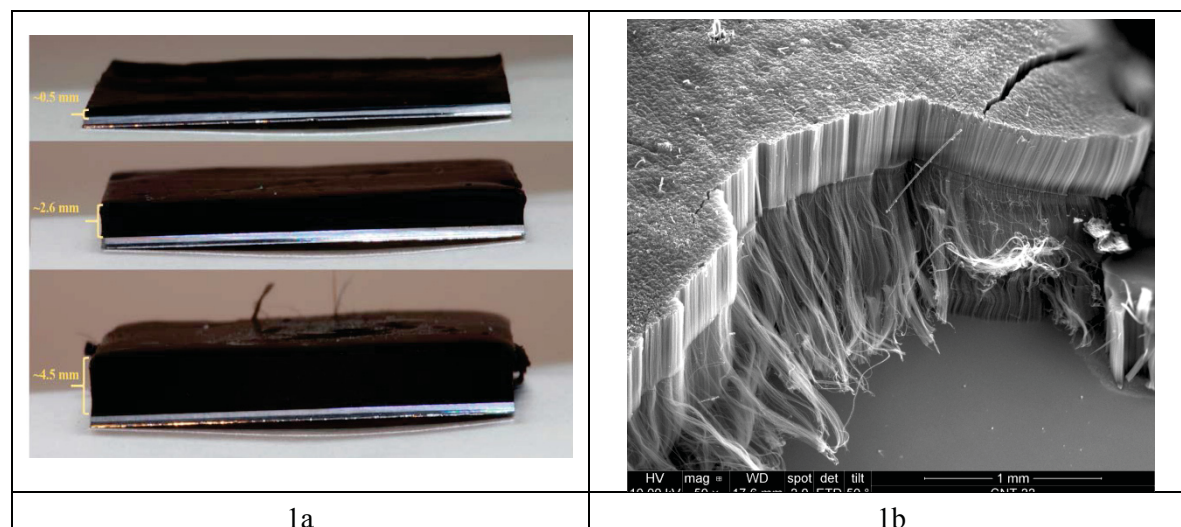
### Benefits to DOE

#### *Technical Relevance*

DOE's mission involves assurance of U.S. security and prosperity through transformative science and technological solutions that address current and future energy, nuclear, and environmental needs. CNT have numerous materials-based applications relevant to DOE's mission, including (1) mixed metal composites for vehicle light weighting, (2) high surface area/permeability catalyst support for improving catalyst efficiency, (3) efficient filtration for removal of biological contaminants from water, (4) enhanced stability of various membrane materials, and (5) sensors for extreme environments.

#### *Innovations and Advancement of the State-of-the-Art*

The collection of in situ gas analysis within the hot zone of a reaction vessel is not a novel concept in and of itself. The use of this technique is rarely applied to the growth of CNTs because the mass spectra of many of the species of interest are complex and the gas environment at high temperatures can be hostile for mass spectrometry. However, the lack of in situ experimental data has severely limited the CNT research community's ability to interrogate the growth mechanism.<sup>1</sup> Collection of gas composition data in situ provides valuable insight into the role of each component within a CNT reactor's influent gas. This insight advances the CNT state-of-the-art by improving our understanding of the reaction mechanism. Ultimately, this will allow better control over the growth of CNTs.



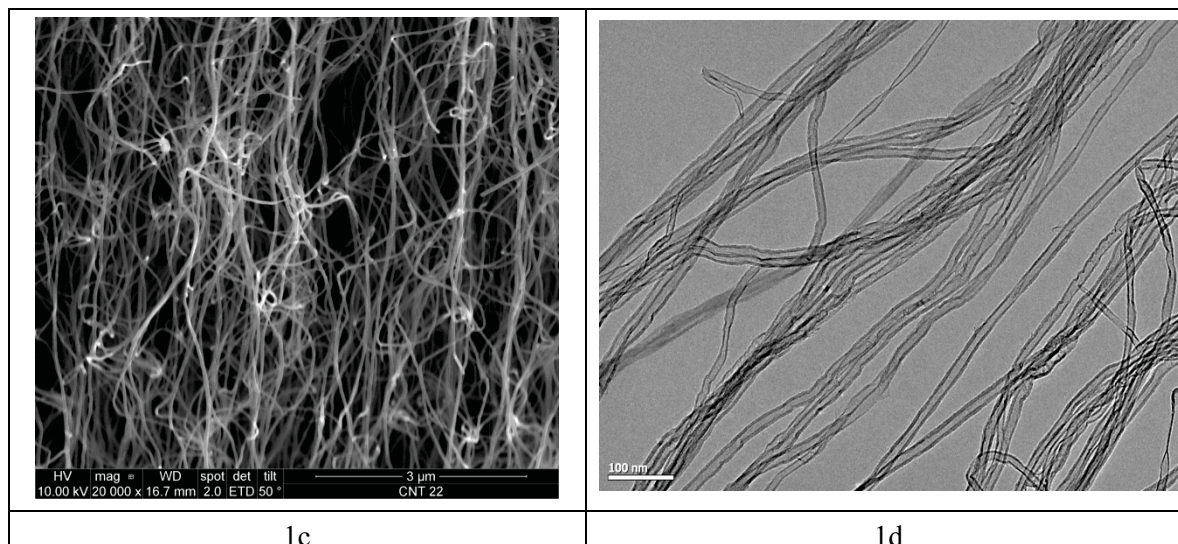


Figure 1. (a) Shows three wafers of vertically aligned CNTs grown under the same conditions, but with different flow rates. (b) Wall of vertically aligned array of CNTs; the arrays are approximately 2.2 mm in height. (c) Magnified view of CNT array. (d) Individual CNTs taken from a vertically aligned array of CNTs. CNTs produced are typically single-walled and have varying amounts of amorphous carbon deposited on the exterior depending on growth time and gas composition.

## Reference

Puretzky, A. A., D. B. Geohegan, S. Jesse, I. N. Ivanov, and G. Eres. 2005, *Applied Physics A*, Vol. 81, pp. 223-240.

## **15-039—Transient Modeling of Integrated Nuclear Energy Systems with Thermal Energy Storage and Component Aging and Preliminary Model Validation via Experiment**

Shannon M. Bragg-Sitton,<sup>1</sup> J. Michael Doster,<sup>2</sup> Stephen D. Terry,<sup>2</sup> Carol Smidts,<sup>3</sup> Qiao Wu,<sup>4</sup> Andrew Klein,<sup>4</sup> Charles Forsberg,<sup>5</sup> and Patrick McDaniel<sup>6</sup>

### **General Project Description**

Nuclear power is proposed to offset intermittent renewable generation sources via tight, behind-the-grid coupling to provide clean energy to the electricity, industrial, and transportation sectors. Thermal and electrical energy storage technologies are also investigated for inclusion in integrated energy systems to maximize reactor utilization and flexibility. Steam diversion in a nuclear hybrid energy system to thermal storage or other applications will initiate a reactor transient in a tightly coupled system. Validated modeling tools are required to adequately characterize coupled system response in combination with operational strategies, control algorithms to optimize for transient maneuvers, and to address the potential impact of degrading component health. This project includes modeling of energy storage systems integrated with nuclear systems, modeling of anticipated transient behavior/response of subsystems, conducting limited experiments and mining existing data to validate these models, and assessing the impact of aging and degradation of key components (e.g., valves) on system operation.

### **Summary**

This project leverages university partner expertise and access to previous research, with management and coordination provided by the INL principal investigator. Overall scope focuses on transient modeling of integrated, hybrid energy systems that would incorporate nuclear and renewable generators, electricity production, an industrial process, and energy storage. Preliminary investigation of advanced energy conversion systems that could be coupled with advanced reactor technologies as an evolutionary application for nuclear hybrid energy system was also included in FY 2015 via MIT and UNM partners.

NCSU developed detailed models for an integral pressurized water reactor (IPWR) and for selected energy storage systems. An existing NCSU IPWR model was modified for hybrid applications, allowing simulation of baseload and load-following operation and diversion of steam and electrical power during off-peak electrical demand. Model variations include forced convection (complete) and natural circulation (in testing). In each scenario, control strategies have been implemented to modulate bypass flow such that turbine electrical demand is satisfied and the reactor runs at approximately 100% nominal full power. NCSU completed a model for chilled water energy storage using both electric and absorption chillers. A computational fluid dynamics model of the storage tank illustrates charging and discharging behavior. Absorption cycle modeling considers transient effects, which has not been undertaken previously. Work to couple the thermal energy storage and IPWR models to provide a representation of the entire system is continuing. A model for a sensible heat thermal storage system using thermal oils was also created. This system uses excess reactor steam to heat a fluid, which is stored in a hot tank. The tank charging model was coupled to the IPWR model to establish reactor and steam control valve settings and to determine the ability of the two systems to work together. Modeling of compressed air energy

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<sup>1</sup> Idaho National Laboratory

<sup>2</sup> North Carolina State University

<sup>3</sup> Ohio State University

<sup>4</sup> Oregon State University

<sup>5</sup> Massachusetts Institute of Technology

<sup>6</sup> University of New Mexico



storage with an open Brayton cycle system is complete with a variety of turbine input pressures and temperatures possible, plus consideration for inclusion of a recuperator. A generated table of heat rates indicates that off-line compression of the air can achieve significantly higher power outputs with lower carbon production. Steam accumulator modeling is not yet complete.

NCSU is also developing a model of the Oregon State University multi-application light water reactor (MASLWR) scaled thermal-hydraulic test facility for benchmarking studies. Oregon State University has mined data previously collected at MASLWR for use in model validation. Available test data include steady operation (using natural circulation) at various core powers, transition between power levels, and a loss-of-feedwater accident, including reactor blowdown, containment condensation, and cooling pool heat exchange. Scaling of earlier facility models has been completed to match the modified facility layout, and load-following simulations have been conducted. The RELAP5-3D model for the MASLWR facility was also completed in FY 2016, with a more formal validation of results currently underway using the “SP-3” power ramping test conducted in 2010.

OSU tasks include (1) evaluation of the impact of system duty cycles on aging, control, and reliability of the steam bypass and steam delivery system within an integrated hybrid energy system and (2) leveraging OSU distributed test facilities to determine the probability of a remotely distributed test remaining stable. OSU is applying a two pronged model-based approach to design an online monitoring system, with low-level component models using parametric models of aging and duty cycles, and high-level system models using qualitative fault propagation methods through the secondary loop system. OSU completed the initial low-level model of bypass valve failure that accounts for aging and increased duty cycles that would be imparted in a hybrid energy system configuration. The fault propagation approach (i.e., integrated system failure analysis) was carried out on a simplified system to simulate the various failure propagation paths. Tests indicate that integrated system failure analysis can discriminate most hardware and software failure modes. OSU researchers developed an initial model of the secondary loop, including a qualitative model of component failure modes for the condenser and the high and low-pressure turbines, and initiated fault propagation to determine the adequacy of sensors for fault detection and diagnosis. OSU also improved the time series model for network latency to predict control system stability for a specific distributed facility configuration. The task allocation method for switching control software to a potentially more stable route has also been refined.

## Benefits to DOE

This work provides enhanced dynamic modeling capability for integrated, non-emitting energy systems that could play a major role in future U.S. energy planning and development to achieve deep decarbonization goals established in the 2015 Paris Accord (COP21 meeting). Data collected for preliminary model validation will be applied to further enhance dynamic system modeling capability, and methods developed will be used to evaluate the probability of control system stability. In addition, the OSU-developed advanced, digital alarm system will allow real-time processing of data from installed sensors in future hardware applications to minimize the potential of component failure. Testing initiated at the OSU distributed test facility will also provide a basis for future multi-laboratory, integrated testing of simulation models and hardware to further advance nuclear hybrid energy system concepts. This project leverages INL and university expertise to build an enhanced capability in energy system design and development, supporting objectives within DOE-NE and DOE Energy Efficiency and Renewable Energy. In the future, work could influence approaches used within the DOE Office of Electricity. The current modeling work and future hardware work will enable more rapid adoption of the proposed hybrid energy system technology by industry.

### Publications

- Guo, Q., M. Pietrykowski, R. Aggarwal, C. Liu, and C. Smidts, “A Distributed Test Facility for Cyber-Physical Systems: Part I. Fundamental Principles,” *IEEE Transactions on Control of Network Systems*, to be submitted.
- Misenheimer, C. and S. D. Terry, 2017, “Modeling Hybrid Nuclear Systems with Chilled-Water Storage,” *ASME Journal of Energy Resources and Technology*, Vol. 139, No. 1, pp. 012002-1 – 012002-9, January 2017, (doi: 10.1115/1.4033858).

### Presentations

- Doster, J. M. and K. L. Frick, 2016, “Coupling of Thermal Energy Storage with Small Modular Reactors,” accepted for publication in *Transactions of the American Nuclear Society, Winter Meeting*, Las Vegas, Nevada, November 2016.
- Frick, K. and J. M. Doster, 2016, “Design of a Thermal Energy Storage System for SMRs,” *Transactions of the American Nuclear Society, Annual Meeting*, New Orleans, Louisiana, June 2016.
- Guo, Q., M. Pietrykowski, and C. Smidts, 2015, “Dynamic Data Transmission Frequency Decision for Distributed Control Systems,” *NPIC-HMIT 2015*, Charlotte, North Carolina, February 23 through 26, 2015.
- Hoover,\* K., Q. Wu, and S. Bragg-Sitton, 2016, “SMR Re-Scaling and Modeling for Load Following Studies,” *ANS Winter Meeting 2016*, to be submitted.
- Ibekwe, R. T. and C. W. Forsberg, 2015, “Firebrick Resistance-Heated Energy Storage: Existing Technology Base,” *Transactions of the American Nuclear Society, Winter Meeting*, Washington D.C., November 8 through 12, 2015.
- Li, H., S. Bragg-Sitton, and C. Smidts, 2016, “Failure Diagnosis of the Holdup Tank System via ISFA Technique,” *ANS Winter Meeting 2016*.
- Mikkelsen, D., C., Chang, S. M. Cetiner, A. L. Qualls, J. M. Doster, and T. N. Dinh, 2015, “Small Modular Reactor Modeling Using Modelica for Nuclear-Renewable Hybrid Energy Systems Applications,” *Transactions of the American Nuclear Society, Winter Meeting*, Washington, D.C., November 8 through 12, 2015.
- Schneider, E. and C. W. Forsberg, 2015, “Variable Electricity from Base-Load Nuclear Power Plants Using Stored Heat,” *Proceedings of International Congress on Advanced Nuclear Power Plants*, Nice, France, May 3 through 6, 2015.
- Stack, D. C., 2015, “Impact of Firebrick Resistance-Heated Energy Storage (FIRES) on Electricity Prices in a Nuclear Renewable Grid,” *Transactions of the American Nuclear Society Winter Meeting*, Washington D.C., November 8 through 12, 2015.
- Stack D. C., 2015, “Improving Nuclear System Economics Using Firebrick Resistance-Heated Energy Storage (FIRES),” *Transactions of the American Nuclear Society, Annual Meeting*, San Antonio, Texas, June 7 through 11, 2015.
- Teeter, L., F. Teng, W. Marcum, J. Kruzic, M. Anderson, and J. Tucker, 2015, “Supercritical Carbon Dioxide System for Materials Corrosion Testing,” presentation at *Minerals, Metals & Materials Society Annual Meeting*, Orlando, Florida, March 2015.
- Zhu, M., R. Aggarwal, S. Bragg-Sitton, and C. Smidts, 2016, “Effect of Aging and Demand for Reliability Analysis of Degrading Components,” *ANS Winter Meeting 2016*.



### Completed Theses

- Frick, K., 2016, *Coupling and Design of a Thermal Energy Storage System for Small Modular Reactors*, NCSU Department of Nuclear Engineering, August 2016.
- Guo, Q., 2015, *A Distributed Test Facility for Cyber-Physical Systems*, Ph.D. Dissertation, Department of Mechanical and Aerospace Engineering, Ohio State University, Columbus, Ohio, August 2015. (submission to STIMS pending patent application)
- Nickels, J., 2015, *Optimization of Turbine and Heat Exchanger Stages For the Expansion Process of a Compressed Air Energy Storage System*, NCSU Department of Mechanical and Aerospace Engineering.
- Odeniyi, A., 2016, *Hybridization of a Gas Turbine for Compressed Air Energy Storage*, NCSU Department of Mechanical and Aerospace Engineering, July 2016.
- Zhu, M., 2016 *Design of an Online Monitoring System for Degrading Hardware Components Using Failure Modes and Effects Analysis*, Undergraduate Honors Thesis, Department of Mechanical and Aerospace Engineering, Ohio State University, Ohio, April 2016.

### Invention Disclosures, Patents, Copyrights

Invention Disclosure Reference Number T2016-220, “Distributed Test Facility for Digital Instrumentation and Control Systems,” submitted to the Technology Commercialization Office at Ohio State University.

### Interns and Postdocs

Interns: Corey Misenheimer, Konor Frick, John Nickels, Adefolaluwami Odeniyi, M. Tucker Daniels, Qingti Guo, Cheng Liu, Michael Pietrykowski, Huijuan Li, Rachit Aggarwal, Muzhi Zhu, Bobak Rashidnia, Kyle Hoover, Lucas Teeter, Ben Adams, Marco Teeter, Bjorn Westmen, Daniel Stack, Nima Fathi, and David Weitzel

### Collaborations

University: Massachusetts Institute of Technology, North Carolina State University, Oregon State University, Ohio State University, and University of New Mexico

## 15-125—Phosphoranimines for Advanced Battery Applications

Eric J. Dufek,<sup>1</sup> Joshua S. McNally,<sup>1</sup> John R. Klaehn,<sup>1</sup> Harry W. Rollins,<sup>1</sup> and Riley Parrish<sup>1</sup>

### General Description

Organic carbonates are the principal solvent component in electrolytes for Li-ion batteries (LIBs). For most applications, these compounds are effective, yet they suffer at high voltages and there are questions regarding their safety and long-term stability. At relatively low temperatures, carbonate electrolytes and the primary salt in LIBs (i.e., lithium hexafluorophosphate [ $\text{LiPF}_6$ ]) start to break down and form products that impact the performance of other materials in the battery. The ability to maintain performance for a wide range of applications relies on components that minimize degradation and improve overall safety of the battery. In addition to performance improvement, there is a need to understand performance and create toolsets that allow better understanding of degradation. The work performed thus far under “Phosphoranimines for Advanced Battery Applications” has focused on routes for enhancing performance and identifying the scientific shortcoming of different electrolyte blends.

Phosphazenes are a class of molecules that contain a phosphorus-nitrogen ( $\text{P}=\text{N}$ ) bond as the backbone. As such, they can be oriented in a host of different conformations, including cyclic systems, linear polymeric systems, and monomeric forms. While most research in phosphazenes has been associated with solid materials, distinct subsets of phosphazene compounds exist as fluids, such as oxidatively stable, fire-resistant lubricants and hydraulic fluids. Among the many advantages of these P-N-containing compounds, are their non-volatility, non-flammability, thermal robustness, stability at high voltages and, lastly, compatibility with a host of different electrode chemistries. At INL, research has looked at phosphazenes as co-solvent and at additive levels ranging from 3 to 30% of the solvent volume. While effective at improving overall safety of LIB electrolytes, the cyclic triphosphazenes are limited in the amount that can be loaded into the electrolyte without increasing viscosity to a point where declining performance is observed. Rather than continue on a path that is limited in its overall loading level and by enhanced viscosity, the present work focuses on a newer member to the phosphazene electrolyte family in phosphoranimines (PAs, Figure 1). As a different class of phosphazenes, PAs are monomeric and have lower molecular weights. This provides a route for reducing viscosity while still providing the adventitious safety aspects of cyclic triphosphazenes.

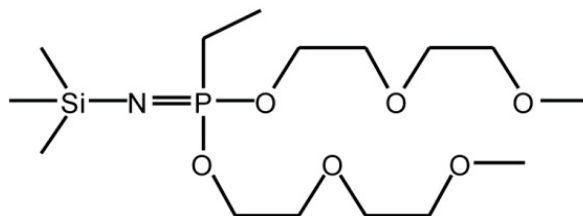


Figure 1. PA-5, a representative PA. The pendants connected to the P are defined as R-groups with the silyl group off the N defined as R.

To-date, the prepared tools have focused on combined computational and experimental activities. On the computational side, focus has been on density functional theory (DFT). Experimentally, work has focused on use of different spectroscopic methods, including nuclear magnetic resonance spectroscopy to identify degradation products and routes for optimizing performance. DFT work has focused on identifying the links between performance and the synthetic route. The link between DFT and synthesis will increase the likelihood of creating new compounds that are successful as battery electrolytes. In parallel with electrolyte development efforts are

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activities associated with creating better ways to evaluate electrolytes with a primary focus on electrolyte stability and electrochemical performance as defined by the nuclear magnetic resonance activities and electrochemical evaluations.

## Summary

Thus far during this project, there has been a peer reviewed publication in *Electrochimica Acta* and multiple presentations at national and international meetings. The work has also resulted in support of three invention disclosures. With regard to the main tasks of this project, progress has been made in the synthetic procedure for phosphoranimine production. Over eight different PAs have been synthesized. These compounds have been characterized using multiple methods, including nuclear magnetic resonance and x-ray crystallography (at the University of Wyoming). Included in the synthesis work has been identification based on DFT modeling of ideal structures for synthesis. Characterization and experimental testing of a PA compound (PA-5, Figure 1) has been carried out and methods for better understanding electrolyte performance in a timely manner have been initiated.

An example of the advanced computational analysis afforded by the use of DFT is exploration of the role that substituents have on the geometric properties of PAs. In particular, the P-N-R' bond angle has been observed to change depending on the nature of the substituent bonded to the N in the PA structure. The change in this bond angle is expected to correlate with the  $\pi$ -bonding character associated with the group and the subsequent bond character of the bond. This bond angle is hypothesized to serve as an important proxy for the nature of PAs as electrolyte materials. For this reason, DFT has helped guide the synthetic goals of this project.

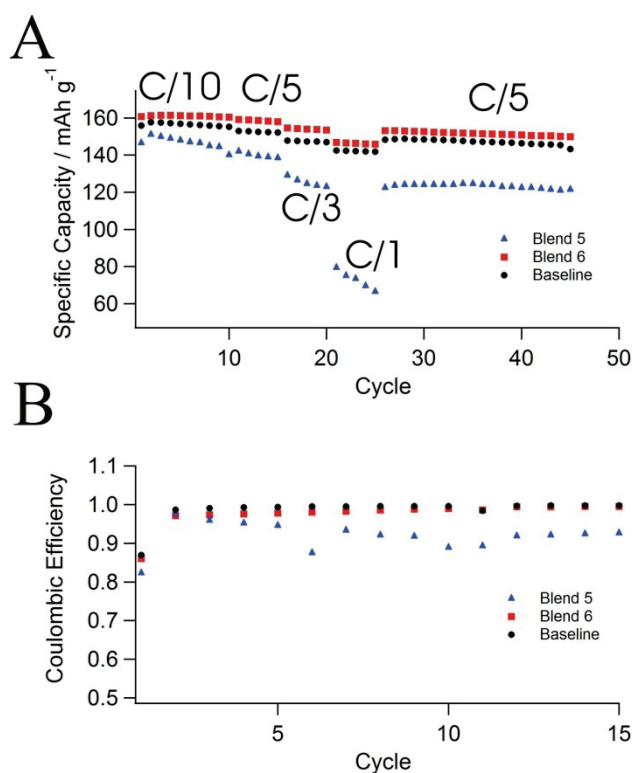


Figure 2. Electrochemical performance data for PA-containing electrolytes with vinylene carbonate as a solid electrolyte interphase forming additive. Blends 5 and 6 contain PA-5.

As highlighted in *Electrochimica Acta*, the initial PAs have decent performance that can be improved based on addition of molecules that enhance solid electrolyte interphase formation. Figure 2 shows there is no distinct

difference in performance between baseline and PA-containing electrolytes. In addition to electrochemical performance, evaluation of the temperature-dependent nature of the electrolytes using differential scanning calorimetry shows that the addition of PAs substantially changes performance and corresponds with an increase in flashpoint for the electrolyte. Ongoing activities with Boise State University have found that inclusion of phosphazenes alters the degradation pathway. As a whole, activities linking DFT, synthesis of new compounds, and development of means for evaluating electrolytes have been successful during the first 2 years of this project. During continuing work, the push to develop more in-depth scientific understanding of the PAs and their role as advanced battery electrolytes will be acquired and new, further refined methods for evaluation will become more uniform.

### Benefits to DOE

Battery manufacturers, DOE, and other government entities such as the Department of Defense have had significant recent interest in new classes of electrolytes and electrolyte additives that improve the safety and performance of LIB chemistries. Improving battery performance is a key concern and a necessity for increasing electrification of the transportation sector in the United States. This move, which is a key focus of DOE, will decrease demand on foreign energy sources and will reduce carbon emissions. From a military perspective, interest in safer electrolytes is tied directly to providing routes to keep soldiers and other military assets safe. Recent discussions with members from the automotive and general battery industries have highlighted the need to more thoroughly validate electrolyte chemistries prior to inclusion in the manufacturing process for batteries. Currently, the deployment process is impeded by the enormous quantity of new electrolyte possibilities and a desire by researchers and inventors to see their contributions succeed. A major focus of industry interest is the ability to perform standardized, high-quality validation that is directly tied to performance-driven metrics. Current research provides a novel electrolyte additive that deviates from the purely carbonate-based line of thinking, which is an approach that DOE is interested in. This project also directly looks at metrics to understand scientific needs for improving battery performance directly in line with the scientific mission of DOE.

### Publications

Dufek, E. J., J. R. Klaehn, J. S. McNally, H. W. Rollins, and D. K. Jamison, 2016, “Use of Phosphoranimines to Reduce Organic Carbonate Content in Li-ion Battery Electrolytes,” *Electrochimica Acta*, Vol. 209, No. 10, pp. 36-43.

### Presentations

Dufek, E. J., J. R. Klaehn, J. S. McNally, and H.W. Rollins, 2016, “Phosphoranimine Electrolytes for Alkali Ion Batteries,” *18<sup>th</sup> International Meeting on Lithium Batteries*, June 2016.

Dufek, E. J., J. R. Klaehn, J. S. McNally, and H. W. Rollins, 2015, “Phosphorus-containing electrolytes – Expansion to include phosphoranimines for Li-ion Batteries,” *228<sup>th</sup> Electrochemical Society Meeting*, Phoenix, Arizona, October 2015.

Parrish, R., 2015, “Electrolyte Performance for Na-ion Batteries,” *2015 Northwest Regional Meeting of the American Chemical Society* (support for hosting a SULI intern from Boise State University).

Smith, K., D. Karsann, R. Parrish, R. Hunt, E. J. Dufek, G. Kamath, S. Sankaranarayana, and H. Xiong, 2016, “Understanding Electrode-Electrolyte Solution Interactions Between TiO<sub>2</sub> Nanotube Electrode and Nonaqueous Electrolytes for Sodium Ion Batteries,” *18<sup>th</sup> International Meeting on Lithium Batteries*, June 2016 (in collaboration with Boise State University).

## **Invention Disclosures, Patents, Copyrights**

BA-892, “Using Hybrid Arrangement of Li-ion and Na-ion Cells to Achieve Better Power Performance in Batteries.”

INL-BA-846, Patent Application No. 14/817,511, “Phosphoranimine Compounds, Electrolyte Solutions including a Phosphoranimine Compound, and Energy Storage Device including Same.”

Patent Application No. 14/720,493, INL: BA-805, “Electrolyte Solutions Including A Phosphoranimine Compound, And Energy Storage Devices Including Same.”

## **Interns and Postdocs**

Intern: Alexander Schoonen

## 15-128—Microstructural Evolution and Mesoscale Coupled Flow-Reaction-Fracturing Processes in Organic Rich Nanoporous Shales

Hai Huang,<sup>1</sup> Earl Mattson,<sup>1</sup> Travis McLing,<sup>1</sup> Darryl Butt,<sup>2</sup> and Paul Leonard<sup>3</sup>

### General Project Description

The project brings together a multi-institutional, interdisciplinary team of computational geoscientists, experimentalists, and material scientists in a focused effort to develop better understanding of the fundamental physics that control the behaviors of organic-rich nanoporous shales and the fluids within them. This project coordinates an array of state-of-the-art characterization and imaging tools, including x-ray, scanning electron microscope (SEM), transmission electron microscope (TEM), and focused ion beam (FIB)-SEM combined with a nano-indentation test to characterize and image the evolutions of the microstructures of nanoporous shales in detail and at the highest possible resolution. The computational component of this project – mesoscale models of coupled diffusion-reaction-fracturing processes – aims to develop multi-physics models that will provide physics-based predictions of microstructure evolutions of organic-rich nanoporous shales induced by changes of fluid chemical compositions, temperature, and pore pressure.

### Summary

During FY 2016, a number of important research milestones, critical experimental and modeling capabilities, and impactful research results were achieved:

1. Successfully applied, the FIB-SEM/TEM imaging and characterization workflow developed during FY 2015 systematically characterized and imaged the nanostructured shale samples provided by the industrial collaborator. One important finding is that the organic matter in the rock matrix exhibits large heterogeneity, even at the nanometer scales. Another important finding is that there is poor connectivity among organic pores (at the highest possible SEM/TEM resolutions).
2. The modeling team successfully applied a reactive force field molecular dynamics model to successfully build realistic kerogen cross-linked polymeric molecule models, which revealed a great amount of subnanometer porosities that could potentially transform our perception of in-place resource estimates for fundamental understanding of transport mechanisms in shales.
3. The modeling team applied a grain-scale discrete element model to predict the deformation and microfracturing of the shale matrix induced by the swelling stress of kerogen and clay due to adsorption and/or water imbibition.

### Benefits to DOE

Research into areas such as microstructural changes of nanostructured, organic-rich shales coupled with fluid flow and volume-changing/stress-generating processes such as chemical transformations, swelling/shrinking, sorption/desorption, and thermal expansion/contraction, is of strategic importance to the nation's energy security and environmental sustainability. These are basic science areas that promise new, scientific discoveries with high economic and societal impact. The research has important implications for sustainable and environmentally friendly recovery of shale gas and oil, geological storage of carbon dioxide in depleted shales, and underground storage of nuclear waste and other hazardous waste.

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<sup>1</sup> Idaho National Laboratory

<sup>2</sup> Boise State University

<sup>3</sup> Pioneer Natural Resources



## Publications

### **Conference Papers during FY 2016:**

Zhou, J., H. Huang, and M. Deo, 2016, “Numerical Study on the Critical Role of Rock Heterogeneity in Fracture Generation,” *50<sup>th</sup> US Rock Mechanics/Geomechanics Symposium*, Houston, Texas, June 26 through 29, 2016.

Zhou, J., H. Huang, and M. Deo, 2016, “Simulation of Hydraulic and Natural Fracture Interaction Using a Coupled DFN-DEM Model,” *50<sup>th</sup> US Rock Mechanics/Geomechanics Symposium*, Houston, Texas, June 26 through 29, 2016.

### **Submitted Journal Manuscripts in Review during FY 2016:**

Pawar, G., H. Huang, P. Meakin, and I. Miskovic, 2016, “Molecular Dynamics Simulation and Subnanometer Scale Characterization of Type I, Type II and Type III Kerogen with a General-Purpose DREIDING Force Field,” *Physics of Fluids*, in review.

Pawar, G., H. Huang, and I. Miskovic, 2016, “Modeling pore-Scale Multiphase Fluid Transport in Shale Reservoirs: A Molecular Dynamics Simulation Approach,” *Energy and Fuels*, in review.

Pathak, M., H. Huang, and P. Meakin, 2016, “Application of General Amber Force Field in Modeling Complex Organic Molecule: Kerogen,” *Energy and Fuels*, in review.

### **Manuscripts in Preparation during FY 2016**

Pawar, G., H. Huang, P. Meaki, E. Mattson, and A. Duin, 2016, “Development of Three-Dimensional, Cross-Linked, Realistic Molecular Kerogen Structure,” to be submitted to *Nature Communications* (in progress).

Pawar, G., H. Huang, and A. Duin, 2016, “Evolution of Subnanometer Scale Organic Material Structure,” to be submitted to the *Journal of Applied Physics* (in preparation).

Xia, Y., H. Huang, and P. Meakin, 2016, “A Mesoscale Modeling and Simulation of Multi-fluid Flow in Nano Pores with Dissipative Particle Dynamics,” to be submitted to *Physical of Fluids* (in preparation).

### **Presentations**

Xia, Y., H. Huang, J. Goral, I. Miskovic, and P. Meakin, 2016, “An Integrated Workflow for FIB-SEM Digital Imaging to DPD Mesoscale Multiphase Flow Simulation in Organic-Rich Nanoporous Shales,” proceedings of *8th International Conference on Porous Media and Annual Meeting of the International Society for Porous Media*, Cincinnati, Ohio, May 9 through 12, 2016.

## Interns and Postdocs

Postdocs: Jing Zhou, Gorakh Pawar, and Patrick Price

## Collaborations

University: Boise State University

Industry: Pioneer Natural Resources

## 15-135—Dynamic Simulations for Large-Scale Electric Power Networks in Real-Time Environment using Multiple RTDS

Manish Mohanpurkar,<sup>1</sup> Rob Hovsapien,<sup>1</sup> and Siddharth Suryanarayanan<sup>2</sup>

### General Project Description

This project aims to test and evaluate necessary components for geographically distributed real-time simulation (RTS) using real-time simulators and interconnectivity between different locations based on wide area network. This connectivity will be used to conduct dynamic and transient tests to analyze large-scale power and energy systems. The other purpose of this project is to address existing gaps for modeling and simulation of newer devices (e.g., power converters operating in a seconds-to-minutes time scale) and evaluate the efficiency and effectiveness of the devices. Challenges in predicting transients ( $\mu$ s to s) under increasing penetration of hybrid energy systems consisting of distributed and renewable sources at the regional and national level will also be addressed using this research work.

### Summary

As part of this project, a data communication link INL and its partner laboratory, the National Renewable Energy Laboratory (NREL), is being used to conduct distributed RTS between Real-Time Digital Simulators (RTDS) at INL and NREL. Since the start of FY 2016, focus has been on conducting power-hardware-in-the-loop simulations in a distributed real-time setup and development of prediction-correction techniques for data latency mitigation in geographically distributed RTS. Progress in distributed RTS connectivity between INL and NREL has enabled successful power-hardware-in-the-loop experiments. A linear predictor has been developed for transmission and distribution power system co-simulation. New approaches are under development for data latency mitigation in collaboration with RWTH Aachen University in Germany. These capabilities will enable local and geographically distributed RTS using power-hardware-in-the-loop and controller-hardware-in-the-loop, electrical/thermal/mechanical subsystem co-simulation at different computational time steps, investigate and establish ways for addressing data latencies, and develop power grid network equivalence techniques on geographically distributed test systems. These capabilities have also helped establish stronger ties and collaboration with researchers and scientists from other national laboratories, academic researchers, and utilities for future dynamic and transient simulation research for large power and energy systems.

### Benefits to DOE

The project enables de-risked large-scale deployment of renewable energy and smart grid technologies pertinent to power and energy systems in the future. This is achieved through high-fidelity RTS and testing in real-world conditions and by using resources through collaborations and interconnectivity between geographically distributed experimental facilities.

### Publications

Liu, R., M. Mohanpurkar, M. Panwar, R. Hovsapien, and A. Srivastava, 2016, “Geographically Distributed Real-Time Digital Simulations Using Linear Prediction,” *International Journal of Electrical Power and Energy Systems*, accepted July 2016.

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<sup>1</sup> Idaho National Laboratory

<sup>2</sup> Colorado State University

- Mohanpurkar, M., M. Panwar, S. Chanda, M. Stevic, R. Hovsopian, V. Gevorgian, S. Suryanarayanan, and A. Monti, 2016, "Distributed Real-Time Simulations for Electric Power Engineering," in *Cyber-Physical Social Systems and Constructs in Electric Power Engineering*, The Institution of Engineering and Technology, London, United Kingdom, November 2016.
- Stevic, M., A. Monti, and R. Hovsopian, 2016, "A Theoretical Framework for Interface Design for Geographically Distributed Real-Time Simulation of Power Systems," *IEEE Transactions on Power Systems*, under preparation.
- Stevic, M., M. Panwar, R. Hovsopian, and A. Monti, 2016, "Improved Prediction Methods for Multi-Site Geographically Distributed Real-Time Simulations," under preparation.
- Panwar, M., S. Suryanarayanan, and R. Hovsopian, 2016, "An MCDA-Based Approach for Dispatch of Electric Microgrids," *International Journal of Electrical Power and Energy Systems*, submitted.
- Panwar, M., S. Suryanarayanan, and R. Hovsopian, 2016, "A Performance Metric for Reserve Management in Day-Ahead Dispatch of Electric Microgrids," *Applied Energy*, under preparation.

### **Presentations**

- Hovsopian, R., 2015, "Laboratory of the Future," for the project titled, Dynamic Modeling and Validation of Electrolyzers in Real Time Grid Simulation, demonstration during Dr. David Danielson's INL visit, Idaho Falls, Idaho, June 30, 2015, available: <https://vimeo.com/131928051>.
- Hovsopian, R., 2015, "DRTS-DRTS Link for Distributed Real Time Simulation," presentation as part of U.S. delegation visit, *Aalborg Symposium on Microgrids*, Aalborg, Denmark, August 27 and 28, 2015.
- Hovsopian, R., 2015, "LDRD review," *BEA Board of Science and Technology Committee Meeting*, July 21, 2015.
- Hovsopian, R., 2015, "Dynamic Simulations for Large Scale Electric Power Networks in Real Time Environment using Multiple RTDS," *DOE-NE Review Germantown Meeting*, September 22, 2015.
- Mohanpurkar, M., 2015, "Distributed Real Time Simulations Using RTDS<sup>®</sup>," *RTDS User General Meeting*, San Francisco, California, May 5 through 7, 2015.

### **Interns and Postdocs**

Interns: Julian D. Osorio Ramirez

Postdoc: Yusheng Luo

### **Collaborations**

University: Colorado State University, Florida State University, and Washington State University

Industry: RTDS

National Laboratories: National Renewable Energy Laboratory

## 15-140—Expanding the Utility of Advanced Chemical Physics Models for Electrolytes

Kevin L. Gering,<sup>1</sup> Joshua S. McNally,<sup>1</sup> Eric J. Dufek,<sup>1</sup> and Claire Xiong<sup>2</sup>

### General Description

Electrolytes play a central role in the performance, safety, cost, and life of the systems in which they reside (e.g., batteries, refrigeration cycles, water treatment, and metabolism). Committed resources in time and money are required to obtain laboratory datasets that cover the foremost metrics needed for electrolyte evaluation and optimization. For example, electrochemical energy storage devices (such as lithium [Li]-ion and sodium [Na]-ion cells) are electrolyte-centric and comprise a multi-billion dollar market that is projected to have greater than linear growth in the coming decades, surpassing \$20B U.S. dollars per annum. This collective emphasis calls for fast computational tools that can economically accelerate selection and characterization of advanced electrolyte systems.

The Advanced Electrolyte Model (AEM) developed at INL is a breakthrough in computing architecture that achieves both speed and accuracy in rendering genome-level information on electrolyte systems. This work sought to expand the utility of AEM by coupling it with upfront ab initio techniques such as density functional theory (DFT) and molecular dynamics that would provide reliable initial estimates of key molecular-scale interactions involving configurational and kinetic metrics. This coupling of modeling methods opens new avenues for applying a unique INL capability to support competitive research in areas of interest to INL, DOE, the Department of Defense, academia, and the private sector. Both bulk and in situ electrolyte behavior were studied with the AEM/DFT package, because it is well known that interfacial processes can dominate system performance (e.g., in batteries). The enhanced AEM tool will be used to gain competitive advantage in proposals.

### Summary

#### *Project Objectives*

- Determine the route for combining quantum chemistry technique DFT with chemical-physics-based AEM.
- Use the combined method for new targets (e.g., electrolytes for Na-ion battery systems), providing genome-level property information toward screening and design.

#### *Technical Challenges*

Ab initio/quantum techniques require long simulation times for convergence, yet provide high-fidelity molecular configuration data. Chemical physics methods (such as AEM) are very fast (about  $10^6$  faster than DFT) and use time-averaged configuration quantities. The challenge was to find common interchangeable metrics between the AEM and DFT methods so they could be used in concert.

#### *Novelty*

The resultant capability is a tool for exploration for new and existing electrolytes. It boasts both accuracy and speed, greatly reducing the time and resources needed to characterize these thermodynamically complex systems. While this combination is novel, it could become an industry standard across multiple sectors.

#### *Technical and Market Benefits*

- Competitive research advantage in complex electrolyte systems

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<sup>1</sup> Idaho National Laboratory

<sup>2</sup> Boise State University

- Quick, high-fidelity answers
- Massive output over several property categories
- Industrial relevance across multiple sectors (e.g., batteries and water treatment)
- AEM large-scale optimization can consider millions of unique conditions per run
- Clean energy mission is supported by accelerated/deepened information toward better battery electrolyte systems.

## Results

- The DFT-to-AEM bridge was successfully developed, where DFT information can “prime the pump” for AEM simulations. Energy-minimized solvent-to-ion DFT configurations provide a logical connection to AEM chemical physics inputs. DFT calculations only need to be run once per ion-solvent pair, then the results are used as a reference configuration within the AEM library.
- 15 compounds were added and validated to the AEM library, including sodium salts, INL phosphazenes, and solvents relevant to high-voltage battery systems.
- IDR BA-892, “Using Hybrid Arrangement of Li-ion and Na-ion cells to Achieve Better Power Performance in Batteries” (Gering, Dufek, and McNally).
- Exploratory simulations compared Na-ion and Li-ion systems and found a lesser extent of ion solvation for Na-ion, resulting in improved transport properties (e.g., reduced viscosity and higher conductivity) and reduced binding energies. This translates to improved battery designs based on optimized Na-ion electrolyte systems. This aspect helped support our collaboration with Professor Claire Xiong at Boise State University.
- Nano-scale electrochemical double-layer simulations by AEM clearly showed that Na-ion systems have double-layer resistivities that are half or less those of comparable Li-ion electrolytes at lower temperatures. It is plausible that Na-ion cells can have improved power over Li-ion at low temperatures, which provides a path forward for resolving a persistent problem with Li-ion systems.
- Two presentations to professional meetings and two manuscripts to peer-reviewed journals.
- Recent private sector interest in AEM capability, both domestically and abroad (e.g., Toyota, A123 Systems, Ceramtec, Mercedes-Benz, Renault, DuPont, Xalt Energy, Dalhousie University, and others).

Figures 1 through 5 give examples of approach, novelty, and modeling results that yield insights into key property predictions. This capability gives INL a unique advantage in timely, thorough, and economical exploration of complex electrolyte systems at unparalleled computational speeds. This can be one basis for competitive proposals.

## Benefits to DOE

Overall, the outcomes from this work support DOE Clean Energy initiatives, particularly in cold-climate regions. In relation to optimizing the value to DOE investment in battery energy storage, the outcome of this work brings higher confidence and rigor to the design and characterization of advanced electrolytes and related materials for batteries. This work has application to the electrical grid and other stand-alone battery energy storage systems and aligns with DOE Office of Energy mission areas through the Office of Electricity Delivery and Energy Reliability. Other potential contributions exist within electrified vehicle battery development under the DOE Vehicle Technologies Office and the private sector tied to DOE programs.

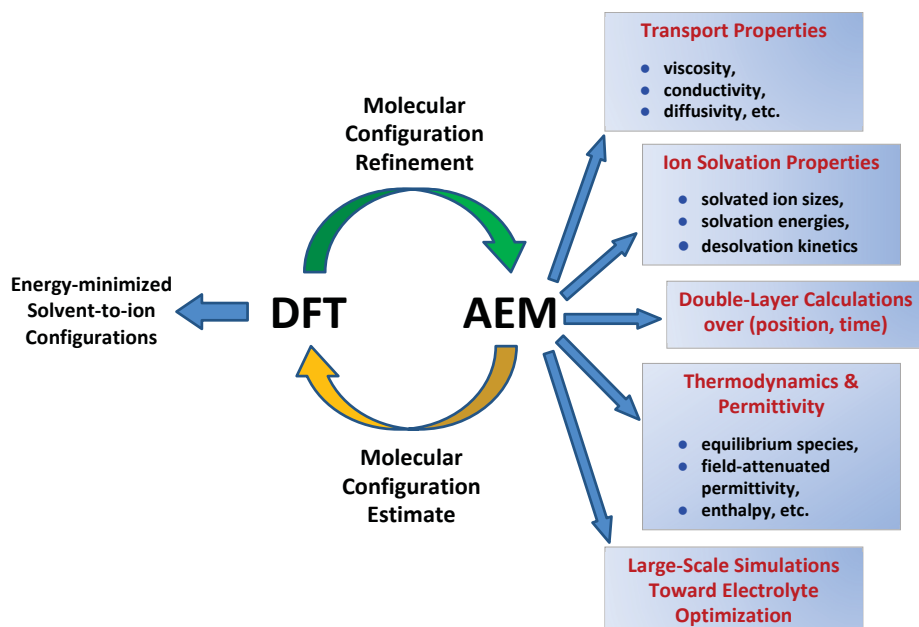


Figure 1. Approach and novelty for combining AEM and DFT methods. A strong synergy is gained by exploiting DFT accuracy for solvated ion structures with the inherent speed of the chemical physics AEM.

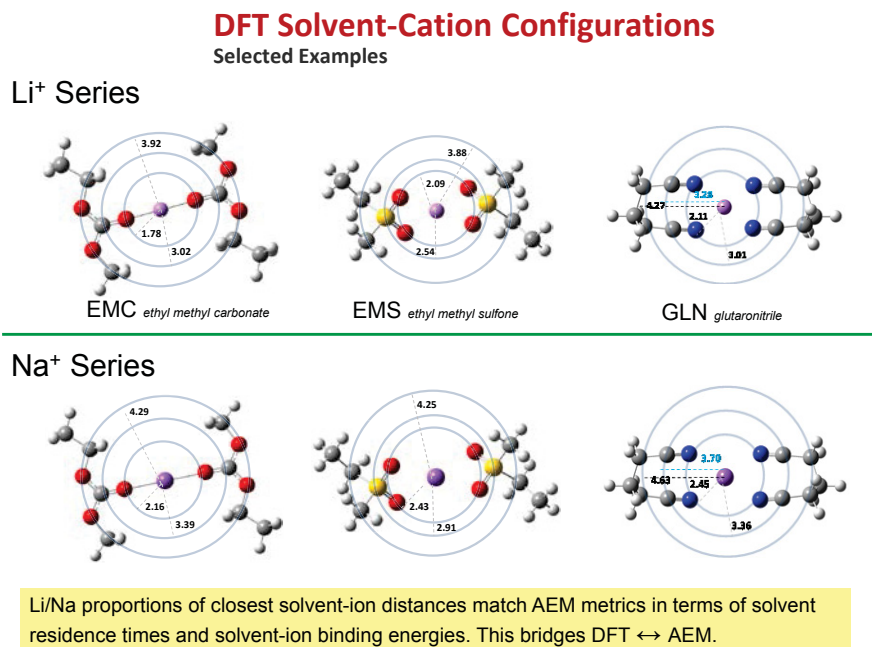


Figure 2. Examples of DFT energy-optimized, solvated ion structures for lithium and sodium cases, using different classes of solvents. The key metric is the stand-off distance between an ion and the volume centroid of the solvent at the energy minimized configuration.



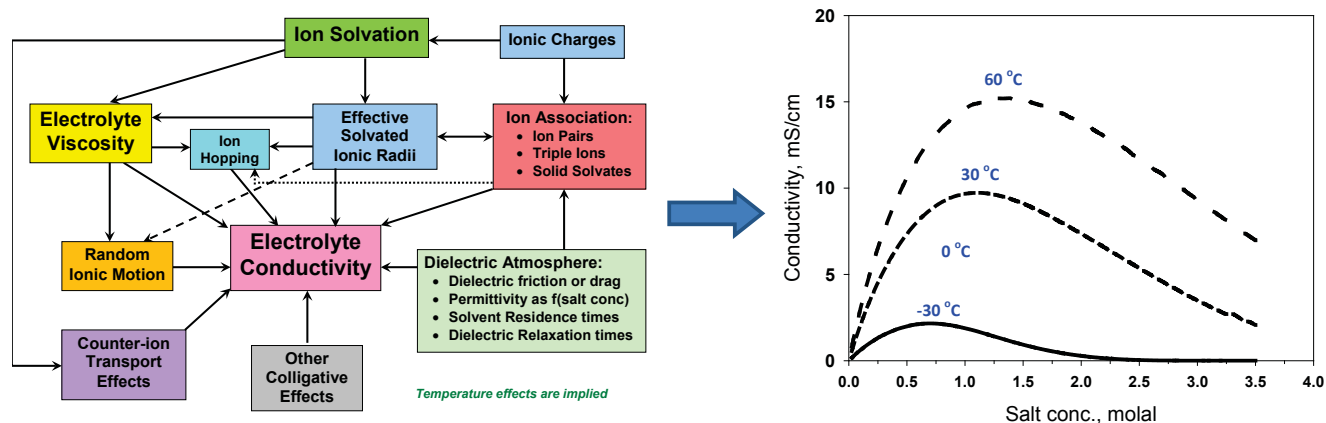


Figure 3. Example of model complexity related to one important property—conductivity. DFT inputs arrive at the “Ion Solvation” block.

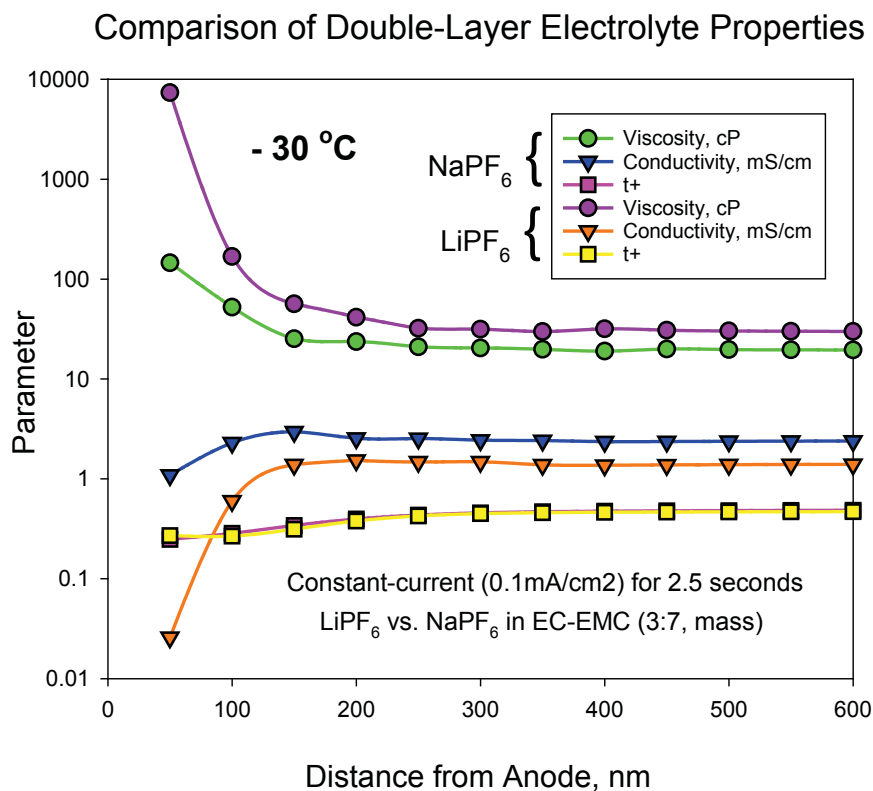


Figure 4. Comparison of calculated electrolyte double-layer properties at low temperature for lithium versus sodium based electrolytes, using nano-scale electrochemical double-layer simulations by AEM (here, on the anode side). An advantage is seen for the Na-ion system due to lower viscosity and higher conductivity during cell operation.

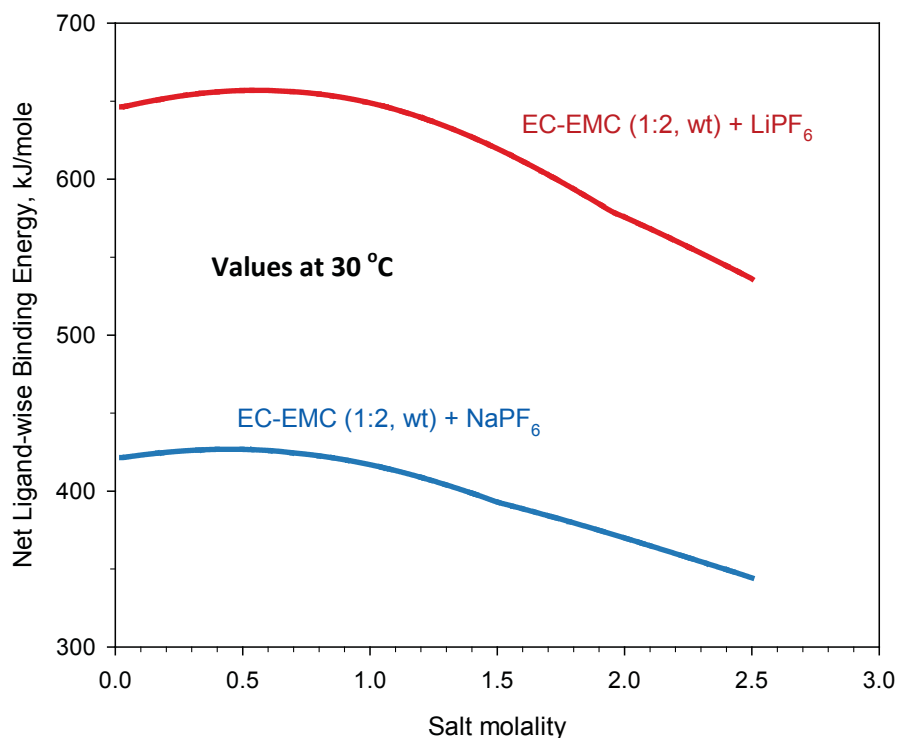


Figure 5. Comparison of solvent-to-cation binding energies, looking at Li-ion versus Na-ion systems. The Na-ion case shows about one-third less binding energy, giving an advantage for interfacial processes in electrochemical cells that require cation desolvation during operation.

Collectively, success of this work enhances U.S. domestic energy security (with a related decrease in use of fossil fuels) through battery research and development that is accelerated and centered on state-of-the-art computational techniques. Success in low-temperature modeling work gives insights on how to overcome the fundamental limitations of batteries at cold conditions and helps expand battery usage.

## Publications

- Gering, K. L., 2016, "Prediction of Electrolyte Conductivity: Results from a Generalized Molecular Model Based on Ion Solvation and a Chemical Physics Framework," submitted to *Electrochimica Acta*, August 2016.
- Gering, K. L. and J. S. McNally, 2016, "Ion Solvation Metrics and Predictive Methods based on Contributions from Density Functional Theory and an Advanced Chemical Physics Approach," submitted to *Journal of Solution Chemistry*, September 2016.

## Presentations

- Gering, K. L., 2015, "Exploring Electrolyte Properties and Behavior through a Chemical Physics Modeling Approach," *CAES Materials, Modeling, Simulation, and Visualization Workshop*, McCall, Idaho, May 13, 2015.
- Gering, K. L. and J. S. McNally, 2015, "Integration of Chemical Physics and Ab Initio Models for Application to Li-ion and Na-ion Electrolytes," *228<sup>th</sup> Meeting of the Electrochemical Society*, Phoenix, Arizona, October 11, 2015.

## **Invention Disclosures, Patents, Copyrights**

IDR BA-892, “Using Hybrid Arrangement of Li-ion and Na-ion cells to Achieve Better Power Performance in Batteries.”

## **Interns and Postdocs**

Postdoc: Luis Diaz Aldana

## **Collaborations**

University: Boise State University

## 15-146—Tailoring the Kinetic Function of a Surface through the Electronic Effects of Nanoscale Architecture

Rebecca Fushimi<sup>1</sup>

### General Project Description

Nearly 90% of the industrial processes that provide food, fuel, and consumer products rely on catalysis. Many of today's leading chemical transformation technologies remain highly energy intensive, complex, and expensive. The U.S. shale gas industry has entered a period of sustained low natural gas prices and growing supply. We may now envision a paradigm shift away from the historical chemical process that started with heavy petrochemical feedstocks. This project is motivated by a need to shift the focus of industrial processes to build chemical intermediates from more abundant shale gas resources.

The surface of a solid catalyst orchestrates the making and breaking of chemical bonds in a complex reaction mechanism. While there is an endless variety of catalytic architectures one can create, there are only a few chemical bonds one needs to break/form in the interest of energy transformations: (e.g., C-H, C-C, C-O, and O-H). Specifically this work is focused on selective dehydrogenation of light alkanes. This reaction is the first step in upgrading abundant domestic gas resources to higher value alkenes, which serve as the building blocks of vitally important petrochemicals (such as polypropylene and polyacrylonitrile).

While platinum is an excellent metal for C-H bond activation, it binds too strongly to unsaturated intermediates, leading to formation of coke and deactivation. A general strategy is to alloy Pt with another metal to reduce the bonding strength of unsaturated hydrocarbons. New synthetic advances allow precise engineering of multi-component metal architectures and, when combining two different metals at the nanoscale, a material with emergent electronic properties is created (i.e., new properties that are distinct from the summation of the individual parts). For example, surface electronic states will vary widely when two metals are arranged in an alloy configuration, a core-shell configuration, or an assembly of discrete nanoparticles. This work utilizes a unique, low-pressure transient kinetic technique to understand how nanoscale bi-metallic architectures, namely core-shell configurations of different precious metals (e.g., Pt, Pd, Re, and Au) and base metals (e.g., Co, Ni, and Sn) can be used to direct the activation of light alkanes.

### Summary

This project has established INL leadership in the temporal analysis of products (TAP) transient pulse response technique for capturing microkinetic detail on complex catalytic surfaces. A state-of-the-art instrument was installed in September 2015 and an earlier generation model was acquired in 2016. INL now operates two of three instruments available in the United States, with 17 global installations. TAP technology distinguishes INL with unique capabilities and expertise for studying interfacial chemical reactions at a fundamental level that is amenable to applied materials.

Initial TAP experiments focused on a series of novel Pt catalysts mounted on a phase-pure Mo<sub>2</sub>C nanotube support that was prepared via atomic layer deposition in collaboration with researchers at the University of Wyoming. TAP pulse response characterization using carbon monoxide as a probe molecule indicated carbon dioxide formation in the absence of oxygen; this enabled researchers to study the elementary reaction steps of the water-gas shift reaction. TAP findings were able to distinguish unique active sites on these materials that supported a strain-ligand hypothesis between the Pt domains of different sizes and the Mo<sub>2</sub>C support. In

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<sup>1</sup> Idaho National Laboratory

collaboration with the University of Ghent and Saint Louis University, new theoretical tools were developed for analysis of multi-step elementary reaction mechanisms surrounding these interesting data.

Equipment for synthesis of core shell catalytic architecture was set up following the electroless deposition method that is practiced in only a handful of laboratories. A variety of core domain sizes were prepared using Pd and Ni to support a thin Pt shell. High-angle annular dark field -scanning transmission electron microscopy, with energy dispersive x-ray spectroscopy elemental mapping, confirmed the desired core shell architecture with no Pt deposition on the SiO<sub>2</sub> support (Figure 1).

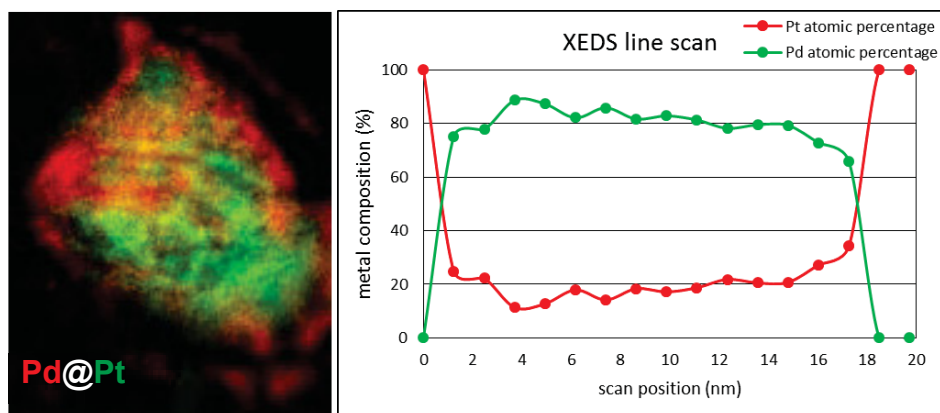


Figure 1. Energy dispersive x-ray spectroscopy elemental mapping (left) and line scan (right) of a 0.49-wt%Pt to 1-wt%Pd/SiO<sub>2</sub> catalyst prepared using the electroless deposition method. Pt is selectively deposited on the Pd metal (i.e., no Pt on the support) and a true bimetallic core@shell structure is detected.

TAP is being used to track changes in ethylene adsorption/desorption and decomposition with respect to metal species, domain size, and shell thickness. These results and trends are compared to theoretical calculations of bond dissociation enthalpies using recently acquired VASP and MedeA software.

## Benefits to DOE

Energy supply plays a key role in our national security. New catalytic technology is needed that can use abundant domestic shale gas resources to replace the conventional production of fuels and chemicals that are presently based on petroleum. This work contributes to fundamental understanding of how catalytic materials can be improved to control a desired sequence of chemical reaction at reduced energy consumption.

## Publications

- Fushimi, R., J. Gleaves, and G. Yablonsky, 2016, "Experimental Methods for the Determination of Parameters," in Z. I. Onsan and A. K. Avci (Eds.), *Multiphase Catalytic Reactors: Theory, Design, Manufacturing, and Applications*, pp. 233-252, John Wiley & Sons ISBN 9781119248477.
- Madix, R., L. Wang, M. Personick, S. Karakalos, R. Fushimi, and C. Friend, 2016, "Active Sites for Methanol Partial Oxidation on Nanoporous Gold Catalysts," *Journal of Catalysis*, accepted for publication.
- Morgan, K., N. Maguire, R. Fushimi, J. Gleaves, A. Gouget, M. Harold, E. Kondratenko, U. Menon, Y. Schuurman, and G. Yablonsky, 2016, "Temporal Analysis of Products (TAP): A Hidden Jewel in the Crown of Heterogeneous Catalysis Kinetic Analysis," *ACS Catalysis* submitted for publication.
- Tan, S., S. Saha, L. Wang, R. Fushimi, and D. Li, 2016, "Experimentally Probing Ligand-Strain Effect via a Novel Catalyst Platform," *Journal of the American Chemical Society* submitted for publication.

Wang, L., C. Friend, R. Fushimi, and R. Madix, 2016, “Active Site Densities, Oxygen Activation and Adsorbed Reactive Oxygen in Alcohol Activation on NpAu Catalysts,” *Faraday Discussions*, Vol. 188, pp. 57-67, 10.1039/C5FD00161G.

### Presentations

Baroi, C., R. Fushimi, and A. Gaffney, 2016, “Techno-Economic and Process Safety Analysis of Ethane to Ethylene Production Process through Oxidative Dehydrogenation (ODH) Route,” *Annual Meeting of The American Institute of Chemical Engineers*, San Francisco, California, November 13 through 18, 2016.

Fushimi, R., E. Redekop, C. Nyapete, J. Gleaves, and G. Yablonsky, 2015, “A Kinetic Fingerprint for Distinguishing Porous Diffusion,” *12<sup>th</sup> European Congress on Catalysis*, Kazan, Russia, August 30 through September 4, 2015.

Redekop, E. A., R. R. Fushimi, G. S. Yablonsky, J. T. Gleaves, and U. Olsbye, 2015, “Temporal Analysis of Products (TAP) – an Advanced Tool for Time-Resolved Kinetic Characterization of Industrially Relevant Microporous Materials,” *15th Norwegian Catalysis Symposium*, Bergen, Norway, December 3 through 4, 2015.

Redekop, E. A., R. Fushimi, and U. Olsbye, 2016, “Kinetic Characterization of Zeolitic MTO/MTH Catalysts with Advanced Temporal Analysis of Products (TAP-3) Instrument,” *XXII International Conference on Chemical Reactors, CHEMREACTOR-22*, London, United Kingdom, September 19 through 23, 2016.

Redekop, E. A., R. R. Fushimi, G. S. Yablonsky, J. T. Gleaves, and U. Olsbye, 2016, “Temporal Analysis of Products (TAP) – an Advanced Tool for Time-Resolved Kinetic Characterization of Industrially Relevant Microporous Materials,” *11th Natural Gas Conversion Symposium*, Tromsø, Norway, June 5 through 9, 2016.

Tan, S., S. Saha, L. Wang, G. Yablonsky, J. Gleaves, R. Fushimi, and D. Li, 2016, “Experimentally Probing Ligand-Strain Effect Via a Novel Catalyst Platform,” *Annual Meeting of The American Institute of Chemical Engineers*, San Francisco, California, November 13 through 18, 2016.

Wang, L., S. Tan, G. Yablonsky, J. Gleaves, D. Li, and R. Fushimi, 2016, “Transient Kinetic Screening of Active Sites and Their Consumption: Pt on Molybdenum Carbide Nanotubes,” *Annual Meeting of The American Institute of Chemical Engineers*, San Francisco, California, November 13 through 18, 2016.

Wang, L., S. Tan, G. Yablonsky, D. Constales, J. Gleaves, D. Li, and R. Fushimi, 2016, “Discriminating Kinetic Pathways with Transient Techniques,” *2nd International Symposium on Catalytic Science and Technology in Sustainable Energy and Environment*, Tianjin, China, October 12 through 14, 2016.

### Interns and Postdocs

Interns: Ken Lim, Shuai Tan, and Christian Reece

Postdocs: Chinmoy Baroi, Soe Lwin, Weijian Diao, and Evgeniy Redekop

### Collaborations

University: Saint Louis University and University of Wyoming



## 16-002—Advanced Carbon Feedstock Processing Using Ionic Liquids

Chenlin Li,<sup>1</sup> Luke Williams,<sup>1</sup> Hongqiang Hu,<sup>1</sup> and Katie Li<sup>2</sup>

### General Project Description

This project objective is to develop efficient coal and biomass conversion technologies. Coal and lignocellulosic biomass are two major carbon-based domestic energy sources with intensified interest for efficient conversion into fuels and chemicals. Current ongoing research focuses on coal liquefaction and biomass thermochemical conversion at high temperature and elevated pressure, developing environmentally benign and low-energy conversion technologies is necessary for addressing the concerns of energy security and process economics. Both coal and biomass have macromolecular network and cross-link structures and contain inhibitory ash and minerals; this, requires efficient depolymerization and demineralization to be further transformed into clean fuels, chemicals, and high-value materials. Ionic liquids (ILs), as a class of novel environmental benign “green solvents,” are receiving more and more attentions. ILs are salts that exist in liquid form at temperatures below 100°C. With their special physiochemical characteristics (such as the low melting point, non-flammability, and negligible volatility), ILs have been recently used as green solvents in many fields. Particularly, research attention has been given to biomass dissolution in ILs for biofuels production; however, application of ILs as solvent, catalyst, or reaction media in coal and biomass depolymerization, upgrading, conversion, product purification, and IL recovery are still limited or in their infancy. The objective of this project is to develop and demonstrate laboratory research and development efforts pertaining to ILs that are based on carbon conversion processes targeting low-rank Wyoming coal and lignocellulosic biomass feedstocks.

### Summary

Technical accomplishments in FY 2016 included the following:

- Performed IL screening studies that targeted sub-bituminous coal and woody pine dissolution and developed and optimized equipment setup and process flow for solid and liquid separation and product recovery.
- Developed methods and performed physio-chemical characterization (e.g., rheology, crystallinity, particle size, and structure) and composition analysis of solid products recovered from coal and pine dissolution.
- Established collaboration work with the University of Wyoming on sample preparation and characterization of the Powder River Basin coal sample and set up membrane separation capability for IL recycling.

Experimental results show that recovered solids from coal and biomass have preserved or increased carbon contents and significantly reduced ash contents in comparison with raw materials; this proves the hypothesis that ILs are efficient solvents to upgrade the biomass and low-rank coal and create clean products by removing ash impurities and improving carbon contents. Figure 1 shows efficient coal and biomass dissolution and dispersion in ILs, and Figure 2 illustrates increased surface roughness and porosity in recovered solids from the IL process.

### Benefits to DOE

This research work aims to enable a fundamentally novel approach to carbon engineering and biomass/coal refining and establish capabilities pertaining to an efficient, low-energy, and environmentally benign IL-mediated process for production of clean fuels and high-value materials. This work supports DOE Office of Energy Efficiency and Renewable Energy’s (Bioenergy Technology Office) mission to transform biomass into fuels and chemicals, as well as the DOE Office of Fossil Energy’s mission to ensure continuous utilization of traditional

<sup>1</sup> Idaho National Laboratory

<sup>2</sup> University of Wyoming

resources for clean, secure, and affordably energy. The efficient utilization of domestic energy resources, such as Wyoming coal and the available billion tons of lignocellulosic biomass, through novel carbon engineering will promote both clean energy and national energy security.

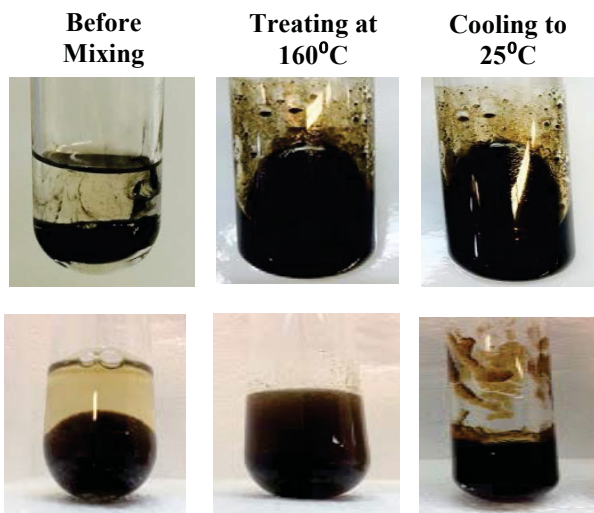


Figure 1. Representative pictures of sub-bituminous coal (top) and woody biomass (bottom) dissolution in ILs.

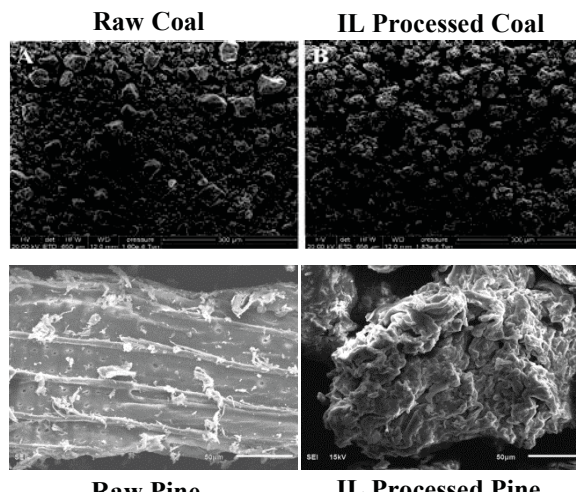


Figure 2. Scanning electron microscopy images showing increased surface roughness and porosity in coal and biomass after the IL process.

### Publications

- Li, C., C. L. Williams, and H. Hu. "Towards the Application of Biomass Ionic Liquid Pretreatment for Biofuels Production – A Technical Review," Invited paper for the Special Issue "Ionic Liquids and Deep Eutectic Solvents for Synthesis, Materials and Energy" *Molecules*, in preparation.
- Li, C., C. L. Williams, J. Allen, B. Thomas, and H. Hu. "Sub-bituminous Coal Dissolution and Dispersion Behavior in Various Ionic Liquids," *Applied Energy*, in preparation.

### Presentations

- Li, C., B. Simmons, and S. Singh, 2016, "Ionic Liquids Processing of Lignocellulosic Feedstocks for Biofuels Production," *Gordon Research Conference – Ionic Liquids*, Newry, Maine, August 14 through 19, 2016.
- Li, C., J. Williams, B. Thomas, and H. Hu, 2016, "Processing Carbonaceous Feedstock Using Ionic Liquids for Sustainable Carbon Engineering," abstract accepted for oral presentation at *2016 AIChE Annual Meeting*, San Francisco, California, November 13 through 18, 2016.

### Interns and Postdocs

Intern: Jared Allen

Postdoc: Shuai Tan

### Collaborations

University: University of Idaho and University of Wyoming.

## 16-176—Development of Direct Carbon Fuel Cells

Ting He<sup>1</sup> and Maohong Fan<sup>2</sup>

### General Project Description

The objective of this project is to develop robust, reduced-temperature direct carbon fuel cells (DCFCs) by applying our expertise in coal engineering, ceramic fabrication, corrosion mitigation, system modeling and integration, and electrochemistry. Upon successful completion of the project, the DCFC technology will offer a new paradigm to the coal industry and achieve a close to zero (operating) carbon footprints without employing expensive carbon capture technologies. This project aims at fundamentally changing the coal industry paradigm from “combustion” to “electrochemical oxidation;” therefore, carbon derived from coal (or biomass or bio-waste eventually) can be used as a clean energy source. The success of this project will impact the regional, national and global coal industry and will also change the way we think of energy and our energy future.

### Summary

This project focuses on developing robust, reduced-temperature DCFCs by integrating our expertise in coal or carbon engineering, ceramic fabrication and characterization, corrosion mitigation, and system modeling and integration with novel perovskite-carbonate composite electrolytes. A systematic approach has been implemented to address materials and process aspects of green coal utilization technology. The proposed research encompasses three synergistic thrusts: (1) conversion of coal to DCFC-grade carbon and high-value chemicals; (2) development of reduced temperature DCFCs; and (3) system modeling for carbon life-cycle and techno-economics. During the initial 6 months since the start of the project, we have established capabilities in preparing novel ceramic powders; formulating tapes, pastes, and inks; and fabricating button cells. Figure 1 shows a schematic flow chart of fabricating DCFCs.

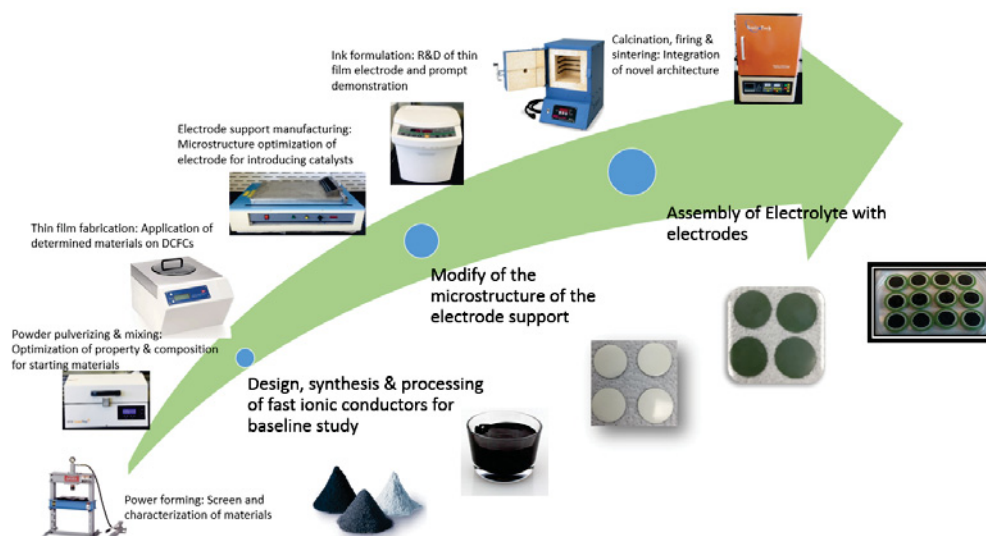


Figure 1. Schematic flow chart of fabricating DCFCs.

<sup>1</sup> Idaho National Laboratory

<sup>2</sup> University of Wyoming

Besides fabrication capability, an electrochemical test stand is built up consisting of a furnace, a gas distribution system, and an electrochemical workstation. A multi-channel potentiostat equipped with electrochemical impedance spectroscopy is capable of running multi-programs such as I through V scanning, potential sweeping, and long-term operation at certain voltage or current density for fuel cells. The test stand can also provide quick feedback of new materials screening when coupled with an in-house designed reactor. Figure 2(a) shows comparison of overall conductivity of our first batch of materials (i.e., doped ceria with conventional electrolyte materials, yttrium-stabilized zirconia tested at our laboratory between 400 and 750°C). The doped ceria exhibits at least one order of magnitude higher conductivity than the yttrium-stabilized zirconia at temperatures below 650°C, as well as much lower activation energy. Therefore, it was first adopted in our fuel cell as an electrolyte for reduced temperature application. At the same time, the University of Wyoming is modifying current looping pyrolysis for the steam pyrolysis experiment, aiming to convert coal to DCFC-grade carbon (low-sulfur) (Figure 2(b)).

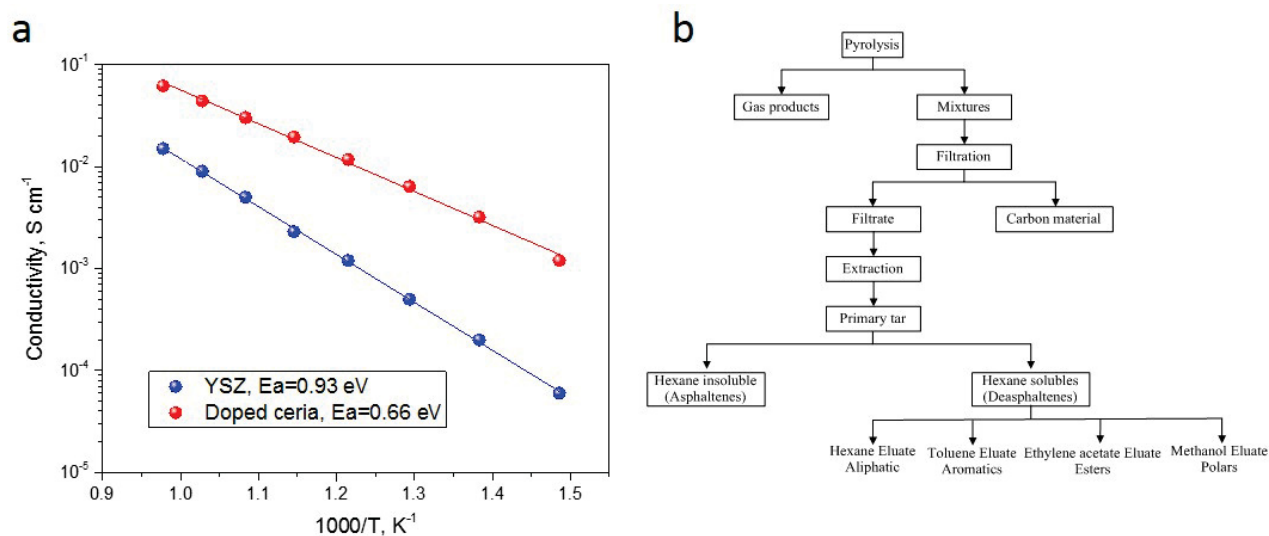


Figure 2. (a) Conductivities of doped ceria and conventional yttrium-stabilized zirconia using a newly built electrochemical stand and in-house designed reactor between 400 and 750°C; (b) steam pyrolysis looping for DCFC-grade carbon from Wyoming coal.

## Benefits to DOE

Energy security, economic vitality and climate change are challenges facing our nation. DCFC technology developed during this project will play key roles across the board for great energy efficiency and productivity and to American leadership in the innovation world as well. The project will also help address the nation's transformation to sustainable and renewable energy self-reliance. The technology will help regional, national, and global coal and power industries to dramatically reduce the carbon footprint, meet the U.S. Environmental Protection Agency's Clean Power Plan target, and reduce environmental impact.

## Publications

He, T., 2016, "Engineering Platinum Alloy Electrocatalysts in Nanoscale for PEMFC Application," *Energy & Fuels Preprints*, Vol. 61, pp. 378.

Tang, M., T. He, H. Adidharma, G. Tan, and M. Fan, 2016, “Coal-Derived Carbon Materials: a Modern Approach to Unlock Nature’s Complexity and Nurture the Gift from the Past,” *Progress in Materials Science*, submitted.

### ***Presentations***

He, T., 2016, “Engineering Platinum Alloy Electrocatalysts in Nanoscale for PEMFC Application,” invited talk, *251st American Chemical Society National Meeting & Exposition*.

He, T., 2016, “Performance and Durability of PtTi Catalyst for Oxygen Electroreduction,” oral presentation, *PRiMe 2016*, The Electrochemical Society.

### **Collaborations**

University: Boise State University, University of Idaho, and University of Wyoming

## 16-215—Electrochemical Manufacturing Processes

Ting He<sup>1</sup>

### General Project Description

The objective of this project is to develop an innovative electrochemical process to replace the conventional energy-intensive thermal process for manufacturing chemicals, plastics, and transportation fuels from natural gas and natural gas liquids feedstocks. Novel electrocatalysts, ion transport membranes, and electrochemical reactors will be developed to convert methane and ethane to olefins or alcohols at relatively low temperature. The overall energy intensity can be dramatically reduced due to much lower operating temperature and close to unity Faraday efficiency. This project aims at fundamentally changing the petrochemical manufacturing paradigm from widely used “thermal” practices (i.e., fossil energy based) to a “clean energy” regime so renewable energy can be deployed on a large scale and support clean energy manufacturing.

### Summary

This project applies electrochemical science to chemical reactions so selectivity and kinetics can be controlled by electrochemical potentials on electrode surfaces. The technology will shift the reaction (i.e., non-oxidative deprotonative coupling and oxidative dehydrogenation) paradigm from chemical to electrochemical. The concept is completely new and involves (1) design of electrocatalysts for methane and ethane deprotonation and dehydrogenation, (2) development of superionic conductors for oxygen-ions and proton transportation, and (3) system integration for performance assessment, economics, and life-cycle analyses. Consequently, a new and comprehensive electrochemical evaluation platform is essential and will greatly facilitate the high yield of research results and outputs, providing the best feedback for new materials and electrocatalysts research and development. During the initial several months of FY 2016, our efforts focused on establishment of the electrochemical test system and design of the membrane reactors. Figure 1 shows an electrochemical membrane reactor testing system and an electrocatalyst and superionic conductor screening system featuring an eight-channel potentiostat and a two-channel electrochemical impedance spectroscopy (both are fully built up and in operation). The electrochemical membrane reactor testing system is able to monitor the electrochemical performance of membrane reactors and analyze the product compositions when coupled with gas chromatography. The electrocatalyst and superionic conductor screening system is a multi-functional system that can be used for electrical and electrochemical property evaluation for bulk materials (i.e., wafers, pellets, and bars) and half cells with different configurations (i.e., two-electrodes, three-electrodes, four-electrodes, and thin-film electrode) on a controllable gas environment at high temperatures. These testing systems are also compatible with a wide range of reactor fixtures that allow exploration of relevant or new research topics at an intermediate temperature range (e.g., hydrogen production by water splitting, ammonia-based fuel cell, and electrical conductivity relaxation).

### Benefits to DOE

This project aligns directly with DOE missions in process intensification and clean energy manufacturing. Improving efficiency and lowering energy intensity in petrochemical manufacturing will not only reduce greenhouse gas emissions and carbon footprint, but also will increase efficiency of deployed capital investment for a wide deployment of clean energy resources, including nuclear-derived heat. The novel electrochemical processes developed by this project will enable DOE’s position in advanced manufacturing and help advance small modular reactor deployment through the market, particularly manufacturing pull – DOE’s Gateway for Accelerated Innovation in Nuclear initiative.

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<sup>1</sup> Idaho National Laboratory



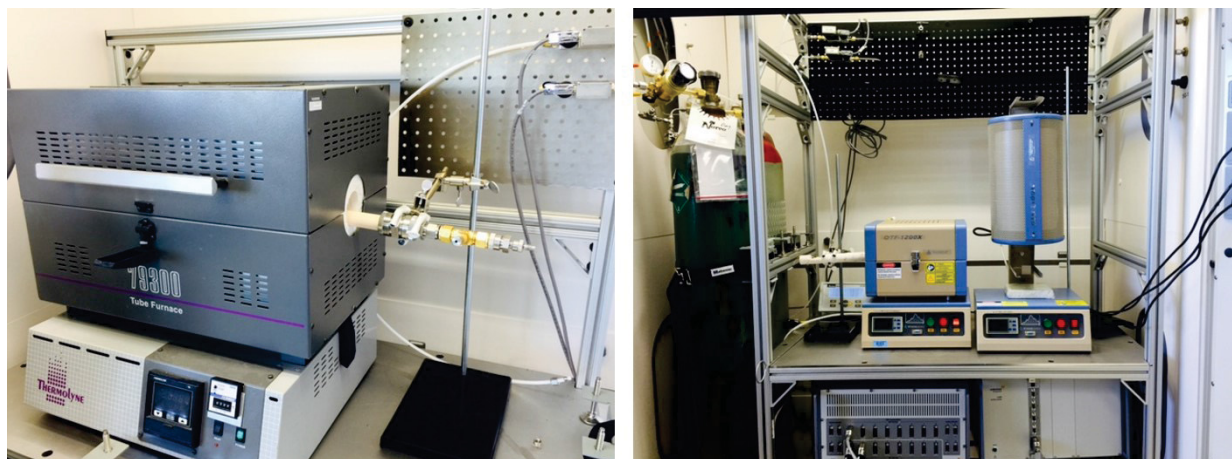


Figure 1. Left: electrochemical membrane reactor testing system and, right: electrocatalysts and superionic conductor screening system.

## Publications

### ***Presentations***

He, T., 2016, "Engineering Platinum Alloy Electrocatalysts in Nanoscale for PEMFC Application," Invited talk *251st American Chemical Society National Meeting & Exposition*.

He, T., 2016, "Performance and Durability of PtTi Catalyst for Oxygen Electroreduction," Oral talk *PRiMe 2016*, The Electrochemical Society.

### **Invention Disclosures, Patents, Copyrights**

IDR# 4104, "Process Intensification for Light Hydrocarbon Gases."

## 16P6-002FP—Kinetic-Based Scale-Up Science for an Energy Efficient Route to Ethylene

Chinmoy Baroi,<sup>1</sup> Soe Lwin,<sup>1</sup> and Weijian Diao<sup>1</sup>

### General Project Description

Ethylene is one of the most important building blocks in the chemical industry and provides the starting point for many essential consumer products (e.g. paints, plastics, and pharmaceuticals). The industrial process presently used in the manufacture of ethylene is enormously energy intensive, inefficient, and expensive. This project focuses on a new, energy-efficient catalytic route to ethylene based on oxidative dehydrogenation (ODH) of ethane. While this route has not been demonstrated industrially, in laboratory studies, the most promising catalysts are based on a complex mixed-metal oxide that is commonly known as M1 catalyst. The overall objective of this project is to initiate an effort for commercializing this M1-catalyzed, ODH-routed ethylene production process. We are looking forward to achieving this goal by enhancing catalytic activity through modification of the M1 catalyst structure. This will be done through novel transient temporal analysis of products (TAP) and operando approaches, testing in a laboratory-scale flow reactor, scaling-up the reaction, and conducting a simultaneous techno-economic analysis of each development stage of the process.

### Summary

The setup for M1 catalyst synthesis has been established and several structures of M1 catalysts have been successfully synthesized (the M1 structure has been successfully identified by the characterization techniques). This exploratory effort will continue into FY 2017. A review paper on the characterization of M1 catalyst has been prepared for submission to a peer-reviewed journal based on our initial work.

A techno-economic and process safety review on the patents for the M1-catalyzed, ODH-routed ethylene production process is underway. Our initial results indicate that the ODH process can be made profitable. An abstract that is based on the techno-economic and process safety analyses study has been accepted as an oral presentation at the upcoming American Institute for Chemical Engineers 2016 conference; the study results will be presented there.

Because the M1-catalyzed, ODH-routed ethylene production process has not been demonstrated on a larger scale, this project will focus on possible modification and upgrading of the M1 catalyst, testing performance in reactors, and scaling-up the catalyst synthesis procedure to enable using these in the large-scale production process into FY 2017. Several research papers are expected to be published based on the results. Also based on the research results of the catalyst upgrading, its performance, and scaling-up, one or two patents might be filed.

### Benefits to DOE

This research project aims to demonstrate a new, energy-efficient catalytic route to ethylene based on ODH of ethane. Upon obtaining successful results, several patents will be filed on both the novel catalyst synthesis and the scale-up procedure. This work will lead to future studies focused on process intensification by utilizing novel reactor designs (e.g., membrane reactor) in combination with a novel catalyst developed at INL. Upon successful demonstration of the technology, INL can be the licensor of this novel catalytic route.

Shale gas is a major source of ethane. The abundant supply and recent decline in shale gas price makes it an attractive source of ethane for ethylene production. The U.S. chemical industry has invested \$15B from 2008 to 2012 in ethylene production, increasing capacity by 33%. As these investments take hold, yielding more supply, the United States could become a major, global, low-cost provider of energy and feedstocks (i.e., ethylene) to the

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<sup>1</sup> Idaho National Laboratory

world-wide chemical industry. Dow Chemical, Chevron Phillips Chemical, and Shell Chemicals are the major ethylene-producing companies in the United States. These companies could directly benefit from this novel catalytic route and would be expected to invest in this research. In particular, this project will demonstrate INL's unique approach to catalyst scale-up based on transient kinetic characterization. We expect to develop Strategic Partnership Projects (SPP) with industry to use our unique capabilities and expertise for similar catalyst scale-up projects.

The catalyst scale-up science outlined in this project matches the goals of the DOE Advanced Manufacturing Office, who has announced an upcoming funding opportunity announcement for process intensification that this work directly complements. Moreover, the fundamental catalyst aspects associated with kinetic understanding derived from TAP will serve as a foundation for future proposals from the DOE Office of Science. Near completion of this project, efforts will be made to harness funding from these DOE agencies.

This work will demonstrate that INL has unique capabilities such as TAP, operando spectroscopy, scale-up, and piloting for catalysis research. The project results can be used to demonstrate how TAP (i.e., a unique facility available at INL) can be used to develop industrial catalysts. This will enhance the reputation of the INL catalyst team and will aid in getting future funding from sponsored programs and industrial companies.

## **Publications**

Lwin, S., C. Baroi, W. Diao, A. M. Gaffney, and R. Fushimi, 2016, "Characterization of the MoVTeNbOx Catalysts during Oxidation Reactions Using In situ/Operando Techniques: A Review" (manuscript in preparation).

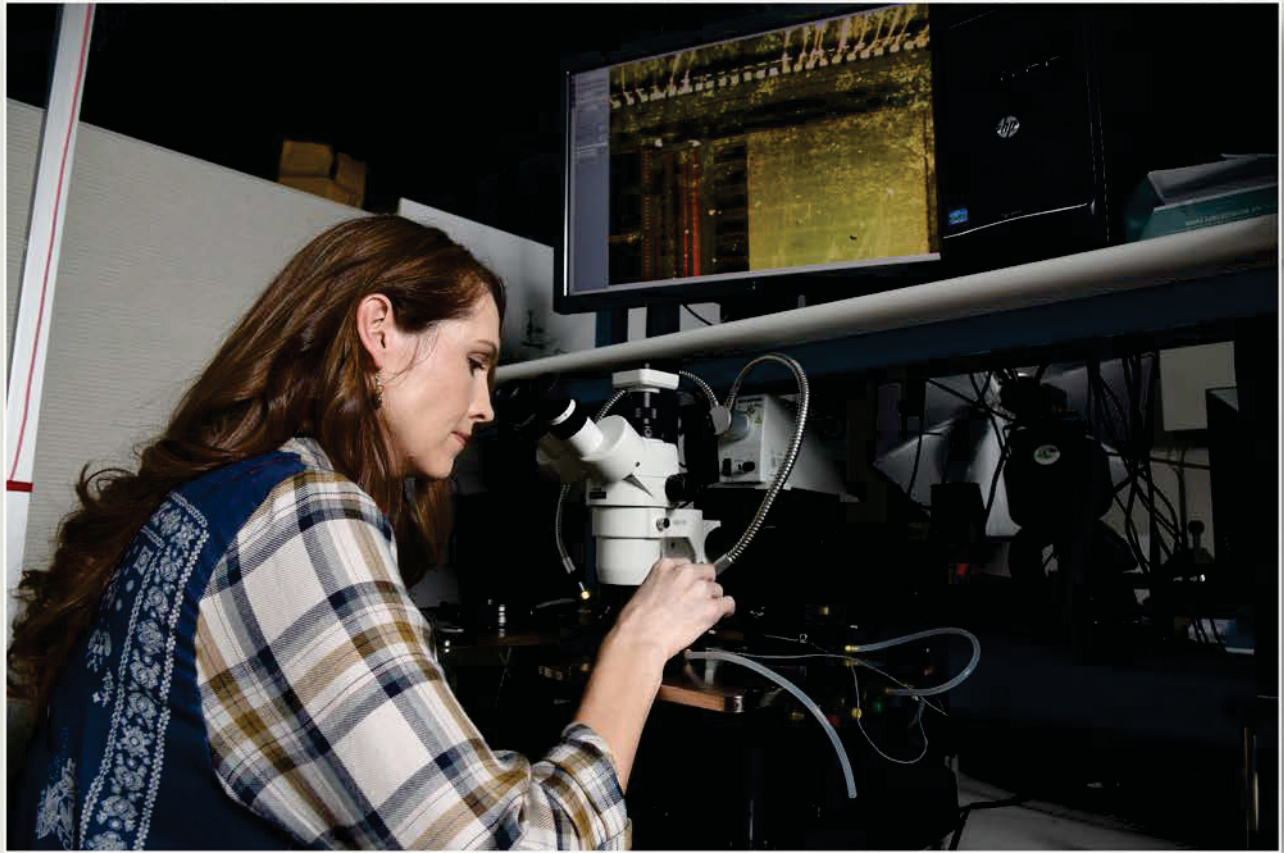
## **Presentations**

Baroi, C., R. Fushimi, and A. Gaffney, 2016, "Techno-Economic and Process Safety Analysis of Ethane to Ethylene Production Process through Oxidative Dehydrogenation (ODH) Route," *American Institute for Chemical Engineers Annual Meeting*, San Francisco, California November 14, 2016.

## **Interns and Postdocs**

Postdocs: Weijian Diao, Soe Lwin





## Securing and Modernizing Critical Infrastructure

14-032—Remote Vulnerability Analysis of CAN Bus Networks  
14-093—All Hazards Critical Infrastructure Knowledge Framework  
14-094—SMC Advanced Armor Materials and Systems Research and Development: Scale-Up Demonstration of Next Generation Armor Steel  
15-083—Visualizing Highly Dense Geospatial Data  
15-096—End-to-End Dynamic Program Analysis for Industrial Control Systems with Concolic Execution  
15-100—Real-Time Process Simulator  
15-111—Adversary Signature Development and Threat Analysis  
16-081—Modeling Thermite Reactions  
16-106—ICS-CAPE: Industrial Control Systems – Cyber Attacks and their Physical Effects  
16-133—Secure SCADA Communications System  
16-152—Wireless Radiofrequency Signal Identification and Protocol Reverse Engineering  
16P6-007FP—Consequence-Driven Cyber-Informed Engineering Methodology Pilot



## 14-032—Remote Vulnerability Analysis of CAN Bus Networks

Jonathan Chugg<sup>1</sup> and Kenneth Rohde<sup>1</sup>

### General Description

The Controller Area Network Bus (CAN Bus) protocol—developed during the early 1980s, first released in 1987, and used in a small number of passenger cars—controls the CAN networks in a modern automobile that may contain 50 or more electronic control units. These electronic control units support a number of different subsystems, including control modules for the engine, powertrain, transmission, anti-lock braking, airbags, power steering, cruise control, door locks, windows, audio system, battery, and charging system. The CAN protocol also supports many different protocols that are used in a wide variety of areas, including automotive, road transportation, rail transport, industrial automation, power generation, maritime, military vehicles, aviation, and medical devices. This research is being performed to discover new technologies capable of enhancing the cyber security of the CAN Bus networks that are used within a typical passenger vehicle, with applications that can also address transportation systems related to aerospace, rail, and maritime.

### Summary

The first year's research focused on accomplishing two goals. The first was to discover if there is an external cyber vulnerability exposure created by exploiting the wireless communications link used in the tire pressure monitoring system to the core CAN Bus network of a modern vehicle. INL research is emphasizing the discovery and subsequent solutions to complex attack vectors that have yet to be publicly disclosed. The second goal was to identify the possibility of circumventing CAN gateways (i.e., modules that shift information between different CAN networks) to migrate from one CAN network to another. Using an attack initiated on a sub-CAN network, research is intended to examine whether an attacker can gain access through CAN gateways to more critical systems.

This vehicle test prototype consists of the electronics of a salvaged vehicle fastened to a display board along with a CAN Bus prototype device that the research team is continuing to develop. This module will be used as a basis for development of a CAN Bus experimental environment to monitor up to three networks and provide alerts on abnormalities. Initial experiments to migrate from one CAN network to another have determined that CAN gateways only allow a limited set of messages to transition across other CAN networks.

The second year of this project, the research team continued to conduct proof-of-concept experimentation to explore new emerging technologies employed in CAN devices across the many transportation sectors. Research included experimentation with new remote wireless and wired capabilities that will soon be employed in the transportation sector, including interconnects between vehicles, charging stations, and the electric grid (Figure 1). Successes include identification of modules responsible during charge and demodulation of CAN signal across the CHADEMO interface.

The third year of research focused on cyber vulnerabilities in vehicle-to-infrastructure communications. Using a modern 50-kW direct current fast charger capable of charging plug-in electric vehicles using either the CHAdEMO or SAE J1772 Combo connector, the team reverse engineered firmware and modules, identified vulnerabilities, developed proof-of-concept demonstrations, and developed scenarios and simulations for cyber attacks (Figure 2).

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<sup>1</sup> Idaho National Laboratory



Figure 1. Overview of vehicle-to-vehicle and vehicle-to-infrastructure.

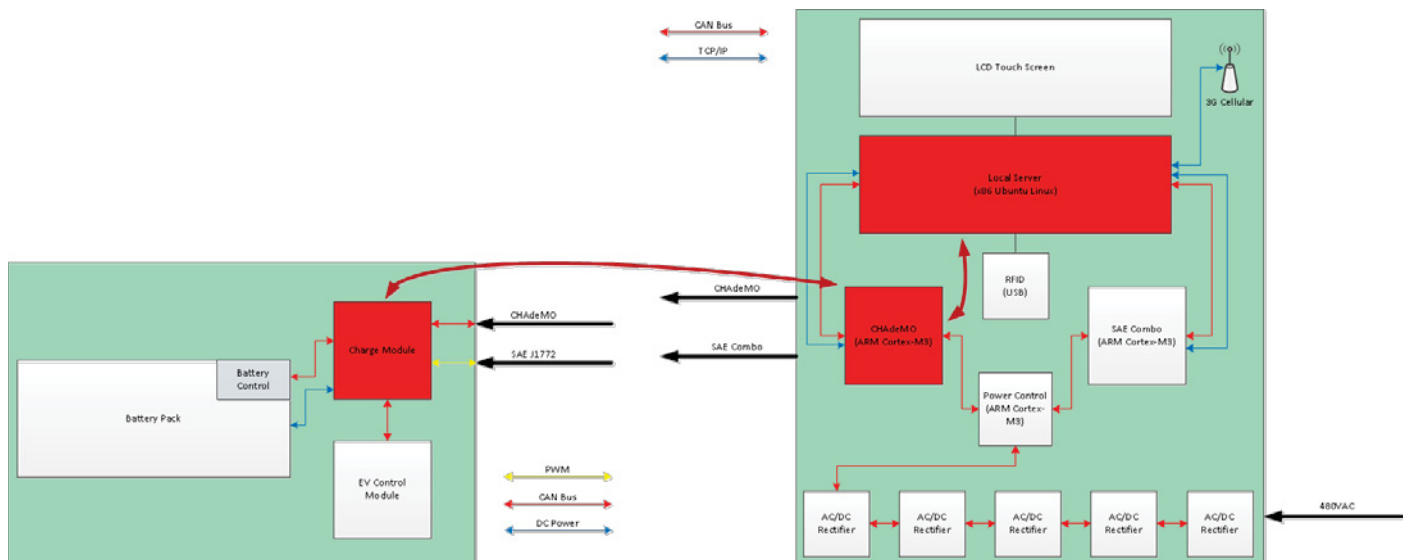


Figure 2. Hypothetical direct current fast charger exploitation scenario.

## Benefits to DOE

The research being performed will enhance cyber security of all sectors of the nation's critical infrastructure that utilize CAN Bus networks, including transportation, law enforcement, energy, and nuclear security. This research initially focused on a typical passenger vehicle and is intended to result in deployed cybersecurity innovations that

are used in all CAN Bus networks. A final goal in the research is to identify the security flaws of an electric vehicle, charging station, and the electric grid and interconnects between all three systems.

This project provided the opportunity to develop the research and refine INL researcher skills and expertise necessary to advance the state-of-the-art research in communications in and around the transportation sector. Ongoing and potential follow-on funding as a result of this research include the following: four operational multi-year and multi-million dollar projects, two multi-year projects that are near a working agreement, and four projects that are in the initial stages of development.

## Publications

Twenty-two presentations and demonstrations were given during the first year, but no publications were developed. The FY 2015 deliverables included one publication at a relevant conference or journal. The following is a summary of notable FY 2014 and FY 2015 presentations:

- Chugg, J. and K. Rohde, 2014, *Strategic Advisory Committee Poster Session*, EIL building at INL, Idaho Falls, Idaho, June 12, 2014.
- Chugg, J., 2014, Battelle Energy Alliance (BEA) S&T Committee, INL, Idaho Falls, Idaho, July 24, 2014.
- Rohde, K., 2014, “Current Cyber Security Efforts,” USCAR: Grid Interaction Tech Team (GITT), ESL Building at INL, Idaho Falls, Idaho, August 2014.
- Chugg, J., 2014, “Vulnerability Analysis of CAN Bus Networks,” Secretary of Energy (Moniz), INL, Idaho Falls, Idaho, August 19, 2014.
- Chugg, J., 2014, “Vulnerability Analysis of CAN Bus Networks,” National Association of Regulatory Commissioners, INL, Idaho Falls, Idaho, August 21, 2014.
- Chugg, J., 2014, “Vulnerability Analysis of CAN Bus Networks,” DOE-NE, Germantown, Washington D.C., August 25, 2014.
- Chugg, J., 2014, “Vulnerability Analysis of CAN Bus Networks,” *Wisconsin Energy Institute*, INL, Idaho Falls, Idaho, September 8, 2014.
- Chugg, J. and K. Rohde, 2015, “Vehicle (In)Security,” *International Atomic Energy Agency International Conference on Computer Security in a Nuclear World*, June 2015.
- Rohde, K. and J. Chugg, 2016, “CAN Bus Security Research Across Multiple Sectors,” *Embedded Security in Cars (escar)*, Idaho National Laboratory, June 2016.

## Invention Disclosures, Patents, Copyrights

IDR BA-814, “CAN Bus Network Safety and Security System.”

IDR BA-858, Rohde, “Instrumentation to Monitor the Physical Properties of Serial Network.”

Provisional Patent 2939-P12608US, “CAN Bus Network Safety and Security System.”

## Collaborations

Industry: USAF Cyber Security Fellows Program

## 14-093—All Hazards Critical Infrastructure Knowledge Framework

Ryan Hruska<sup>1</sup> and Cherrie Black<sup>1</sup>

### General Project Description

The objective of this research project is to develop advanced knowledge discovery and decision support methodologies to improve the nation's capability to conduct infrastructure interdependency analysis in order to better prepare for, protect against, respond to, recover from, and mitigate all hazards. There are two main objectives for this project: (1) development of a knowledge model framework for critical infrastructure and (2) development of adaptive hybrid data and expert-driven algorithms for online learning of infrastructure characteristics.

### Summary

#### ***Knowledge Model Framework***

- Developed a scalable prototype critical infrastructure knowledge framework using a graph database architecture
  - Ingested both unstructured and structured infrastructure information from numerous open source and government-owned sources to generate regional dependency models
- Dependency profiles were generated based on interviews with subject matter experts and reviews of technical manuals for numerous lifeline infrastructures (i.e., water, wastewater, natural gas, electricity, and communications)
- Developed a prototype dependency reasoning model of relationship assignments.

#### ***Text Analytics***

- Refined natural language processing techniques for extracting infrastructure information from unstructured text (i.e., document categorization, named entity recognition, and relationship extraction for critical infrastructure facilities)
- Geolocation tagging for spatial information extraction
- Refined prototype, automated document processing architecture for unstructured text
- Automated population of knowledge framework architecture from unstructured text.

#### ***Visualization***

- Developed prototype knowledge management application to ingest, transform, and visualize critical infrastructure dependency information via a linked graph and geospatial view.

### Benefits to DOE

A major element of the DOE, Department of Homeland Security, and INL missions is to ensure productive, optimal, and secure use of the nation's energy and critical infrastructure resources. This project has benefited INL by developing a scientific understanding of advanced knowledge discovery and decision support methodologies to understand the complex interdependent relationships within these integrated systems, as well as their vulnerabilities to threats and hazards. By developing these methodologies and demonstrating our ability to solve

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these complex analytic challenges, DOE's and the INL's status as a research leader in critical infrastructure protection will be enhanced.

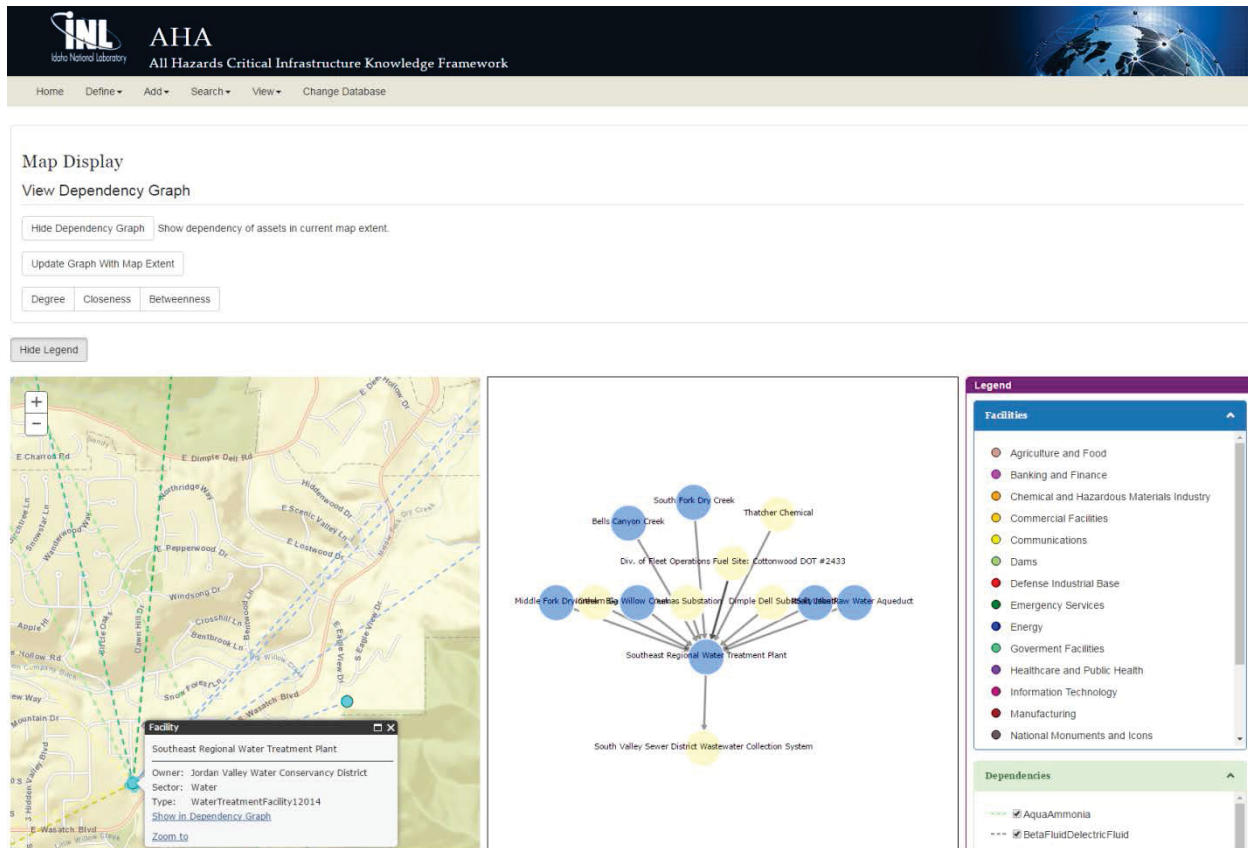


Figure 1. Example dependency model for critical infrastructure.

## Publications

Amarasinghe, K., M. Manic, and R. Hruska, 2015, "Optimal Stop Word Selection for Text Mining in Critical Infrastructure Domain," *IEEE Resilience Week*, Philadelphia, Pennsylvania, August 2015.

## Presentations

Amarasinghe, K., M. Manic, and R. Hruska, 2015, "Optimal Stop Word Selection for Text Mining in Critical Infrastructure Domain," *IEEE Resilience Week*, Philadelphia, Pennsylvania, August 2015.

Hruska, R. and M. Klett, 2014, "Knowledge Framework for Critical Infrastructure Analysis," *IEEE Resilience Week*, Denver, Colorado, August 2014.

Hruska, R., 2015, "Knowledge Framework for Critical Infrastructure Dependency Analysis," *INL/CAES Big Data Workshop: Enabling Collaboration in Scientific Data Analytics*, Idaho Falls, Idaho, May 2015.

Klett, M., 2015, "ICSJWG Emerging Research: Infrastructure Threat Recognition, Resilience and Restoration," *ICSJWG Emerging Research Panel Discussion*, Washington D.C., June 2015.

Hruska, R., 2015, "Knowledge Framework for Critical Infrastructure Analysis," *Department of Homeland Security Dependency Workshop Series*, Idaho Falls, Idaho, Chicago, Illinois, and Washington D.C.

Hruska, R. and S. Staples, 2015, “Natural Language Processing for Critical Infrastructure All Hazards Analysis,” *IEEE Resilience Week*, Philadelphia, Pennsylvania, August 2015.

Hruska, R. and C. Black, 2016, “Critical Infrastructure Dependencies and Community Resilience,” *IEEE Resilience Week*, Chicago, Illinois, August 2016.

### **Interns and Postdocs**

Interns: Maxwell Moseley and Andrew Petti

### **Collaborations**

University: University of Idaho



## **14-094—SMC Advanced Armor Material Systems Research and Development: Scale-Up Demonstration of Next-Generation Armor Steel**

Henry Chu,<sup>1</sup> Thomas Lillo,<sup>1</sup> Thomas Zagula,<sup>1</sup> Colter Angell,<sup>1</sup> and Jeff Anderson<sup>1</sup>

### **General Project Description**

The general objective for the next-generation armor steel (NGAS) is to develop bainite-based armor steels that are superior in performance to those currently available on the market. The motto of NGAS is equivalent performance at reduced weight or increased performance at equivalent weight at a reduced manufacturing cost. A steel alloy with the desired properties was developed during 2009 through 2013 and heat treated in thicknesses up to 1-in. with promising ballistic results. The objective of this project is to produce and heat-treat 2.5-in. thick targets to demonstrate the scalability of the process and the ability to defeat more advanced threats.

### **Summary**

Heat treating was performed in previous years using a benchtop-scale air cooling method. Because of the significantly larger size of the FY 2016 parts, the heat treating process had to be redesigned. The development focus was on scaling the air cooling process that was used in former years. Air flows were simulated using Pointwise and ANSYS Fluent software. The corresponding heat transfer simulations were carried out using SolidWorks 2015. Required cooling air velocity was estimated using this approach and used to design the scaled cooling apparatus.

The NGAS castings were made by a contracting foundry and the final chemical compositions were confirmed by a commercial analytical laboratory. Time - temperature transformation diagrams were generated using software initially developed by researchers at Cambridge University. Calculated cooling curves were overlaid on the time - temperature transformation diagrams to produce a practical continuous cooling diagram for each part.

The time - temperature transformation models predicted that the as-cast chemistries of some of the slabs might not result in greater than 55 Vol% of bainite microstructure formation during the normal air-cooling process. A nitrate-nitrite salt bath was identified as an alternative quench method and a contract was procured to heat treat several of the parts for further analysis and comparison.

Once heat treatment via both salt solution and air-cool quench are completed, it is anticipated the final mechanical properties and microstructural comparison and verification will be conducted in FY 2017 with funding from a research and development customer. Ballistic tests will be performed as final verification of NGAS. All activities had been planned for completion during FY 2016, but some were rescheduled due to supplier difficulties.

### **Benefits to DOE**

NGAS is the result of years of LDRD-funded research into advanced armor technology and represents a potential major departure from the traditional martensitic armor steels that are prone to excessive brittleness, stress-corrosion failure, non-weldability, and limited thickness. NGAS with enhanced performance may be deployed across multiple armored platforms fielded by the Department of Defense. Related research is being conducted into creating conformal composite armor plates using NGAS as a substrate. This has a potential to dramatically improve resistance of body armor against advancing threats. Continued success in these research areas will reinforce the capabilities that INL has developed in materials science research.

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<sup>1</sup> Idaho National Laboratory

## **Invention Disclosures, Patents, Copyrights**

BA-458 1699, “Bainitic Steel Alloy for Armor Applications.”

BA-625 2040, “Cost Effective Steel Alloy for Armor Applications.”

An invention disclosure record is on file and a patent is pending. The formula and process method are Battelle Energy Alliance, LLC proprietary information for official use only.

## 15-083—Visualizing Highly Dense Geospatial Data

Shane Cherry<sup>1</sup> and Robert Edsall<sup>1</sup>

### General Project Description

Analysts, responders, and decision makers who consider the nation’s critical infrastructure (CI) are faced with a staggering amount of data from disparate sources; indeed, our ability to gather data is increasing at a faster rate than our ability to analyze it. Because much of these data are georeferenced, it stands to reason that geographical information system tools can be used as the basis for analysis and visualization of these data in order to convert it to actionable information. However, these tools begin to lose value when the available information becomes so dense it can no longer be easily understood by the human user of the system.

This problem has been a subject of research in the realm of geovisual analytics, which seeks to combine the power of interactive computational mapping and geo-representation technology with decision science and associated research in human cognition (Robinson et al. 2016, Andrienko et al. 2007). Visualization of complex statistical, temporal, and geographic information is seen as a vital component of understanding that information by an analyst, regardless of the domain or context. In this project, we adopt and customize innovative techniques from geovisual analytics to provide solutions to the challenge of comprehending CI-related data vital to homeland security.

### Summary

The majority of the work supported by this project was directed toward solutions for visualization, and associated understanding, of data that are dense and irregular in geographic space and of the relationships among those data points. Specifically, we have investigated methods of constructing an interface that provides simultaneous focus and context of dense irregular point data representing CI facilities.

For example, we introduce a method for a visual, national-scale overview of CI assets, which can number in the millions and be stored in various databases. Traditional techniques for visualizing such dense point sets are plagued by abstruse over-plotting, computational burden, over-resemblance to population density maps, inaccessible raw data, and resulting spurious conclusions. We adapted and incorporated a hexagonal binning method into a web-enabled geographical information system framework, where a geometric tessellation of equally sized polygons is laid over the map and statistical summaries of the assets within each polygon are calculated and represented (Figure 1). Each hexagon can be queried through user interaction and is symbolized in a data-rich way. The underlying map can be panned and zoomed, which is important for analysis because, with these interactions, the position and size of the hexagons relative to the screen remains constant. Therefore, the geographic scale and location of the analysis is constantly updated as the map below is altered. These features allow flexible exploration of the datasets, revealing local detail while maintaining wider-scale context.

Additionally, we developed an alternative, novel, focused-context solution that closely resembles a common real-world activity (thus employing a so-called “interface metaphor”). Our spyglass tool mimics use of a loupe, similar to one used by a jeweler or watchmaker. Upon calling the tool into the interface, a smaller, movable window appears on the cluttered small-scale map and, within the new window, a magnified view of the immediate region in the center of the smaller window (i.e., the “spyglass”) is presented. The spyglass itself can be zoomed in on (the extent of the magnified view is the choice of the user) and can be panned (such that as the user drags the window around the screen, the location that is magnified changes, as it would if using a loupe on a detailed paper map) (Figure 2). This type of tool was previously employed to visualize alternate map layers at the same scale

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<sup>1</sup> Idaho National Laboratory

(e.g., a historical map “underneath” the main map). However, to our knowledge, this is the first implementation that magnifies an existing high-density dataset to focus on local detail.

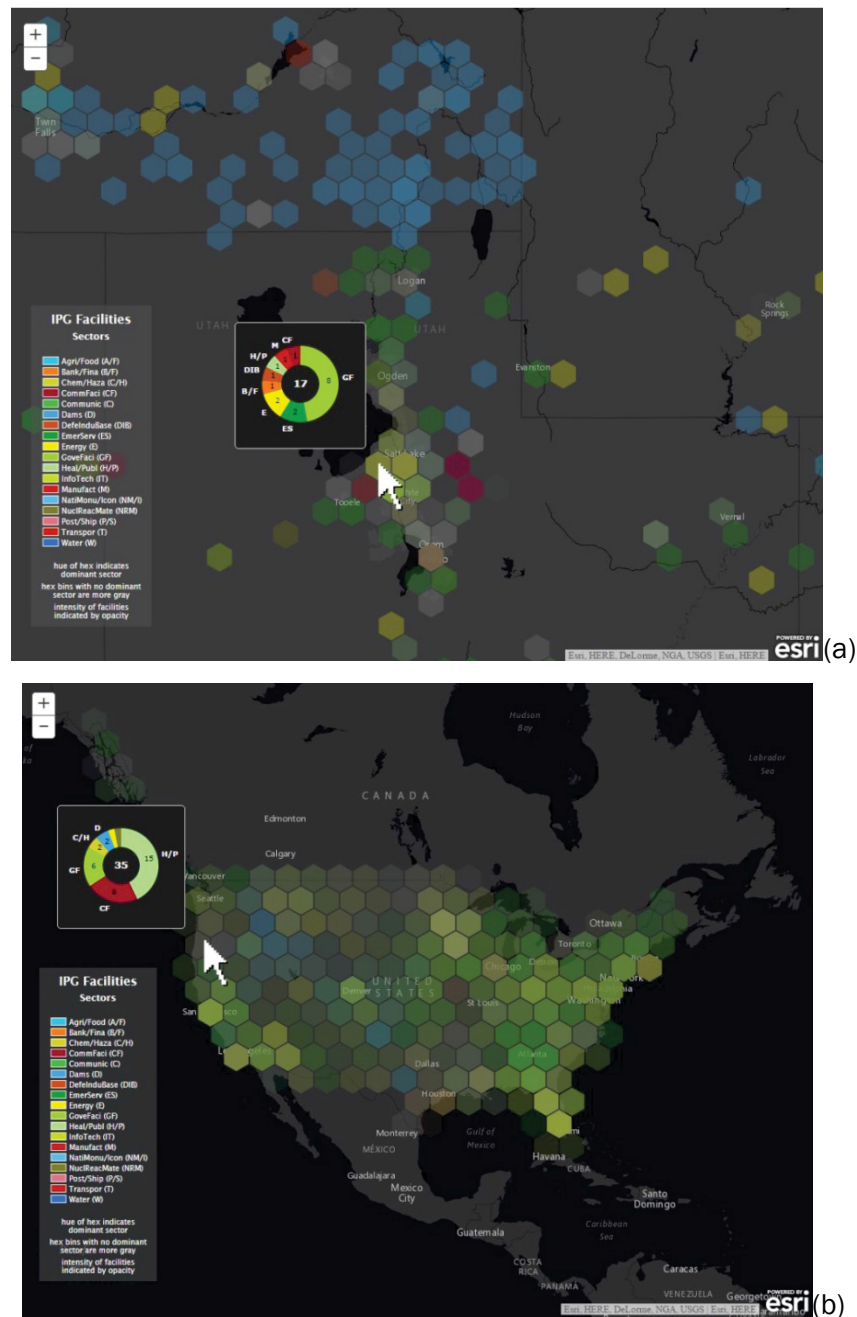
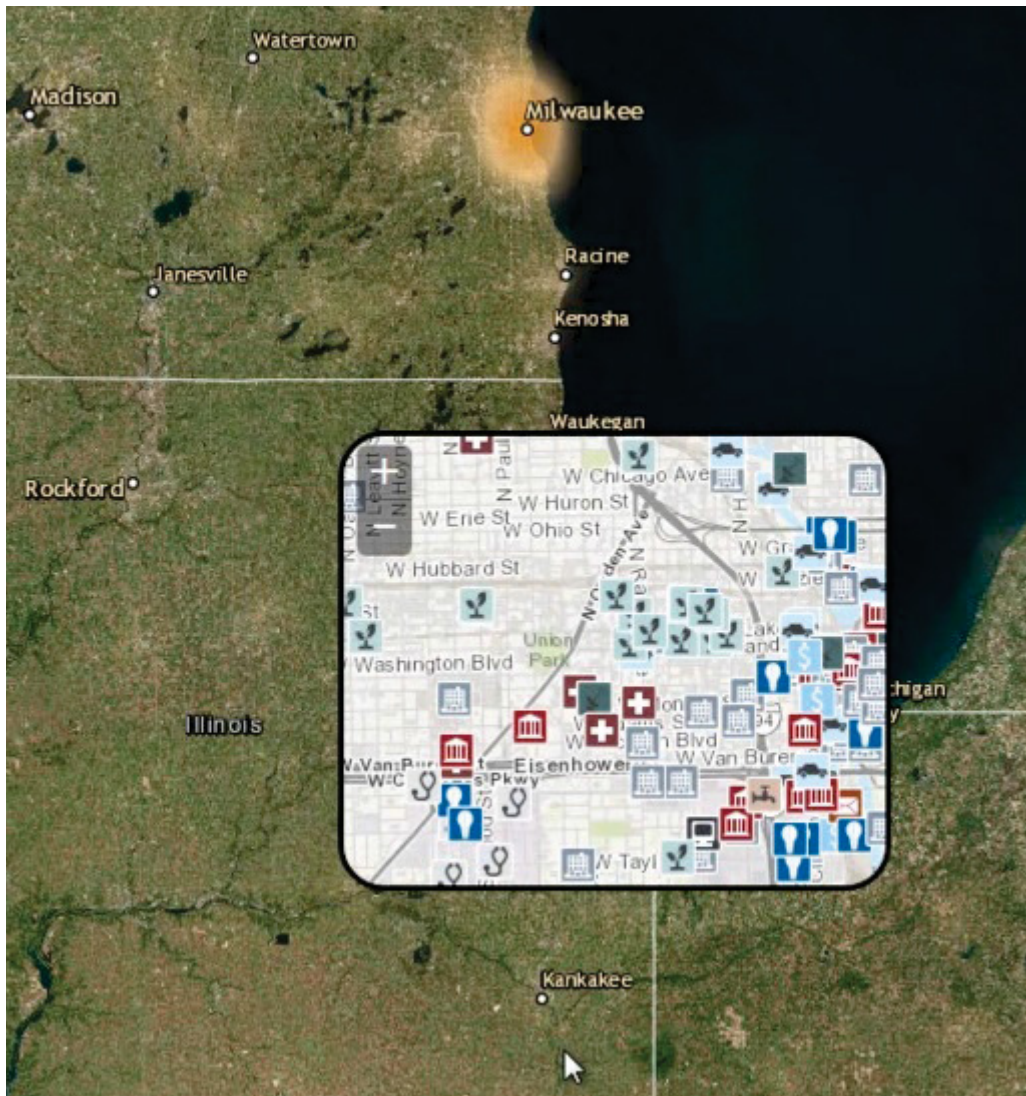


Figure 1. (a) High-density data aggregated to hexagonal bins at a national scale and (b) regional scale re-aggregation showing Department of Homeland Security lifeline sector dominance of CI facilities in each hexagonal areal unit.



*Figure 2. Spyglass tool for examining local detail over a regional overview map. Individual CI assets near downtown Chicago are visible in the spyglass, which has been zoomed to a large relative scale and centered on the regional map over Chicago.*

Finally, our team has developed new capabilities for visualizing the complex dependencies among critical infrastructure assets, which document the cascading effects of a potential natural hazard, cyber/physical attack, or other disabling threat. Our prototype logical dependency graph layout, linked with a geographic representation, can display the strength, direction, and confidence of dependencies among CI assets (Figure 3). While spatial proximity of CI assets often is representative of the importance of the relationships among them, in many cases, essential CI dependencies are physically distant (or even non-spatial). Therefore, “spatializing” networks in a graph that de-emphasizes geographic location can reveal useful and otherwise obscured information.



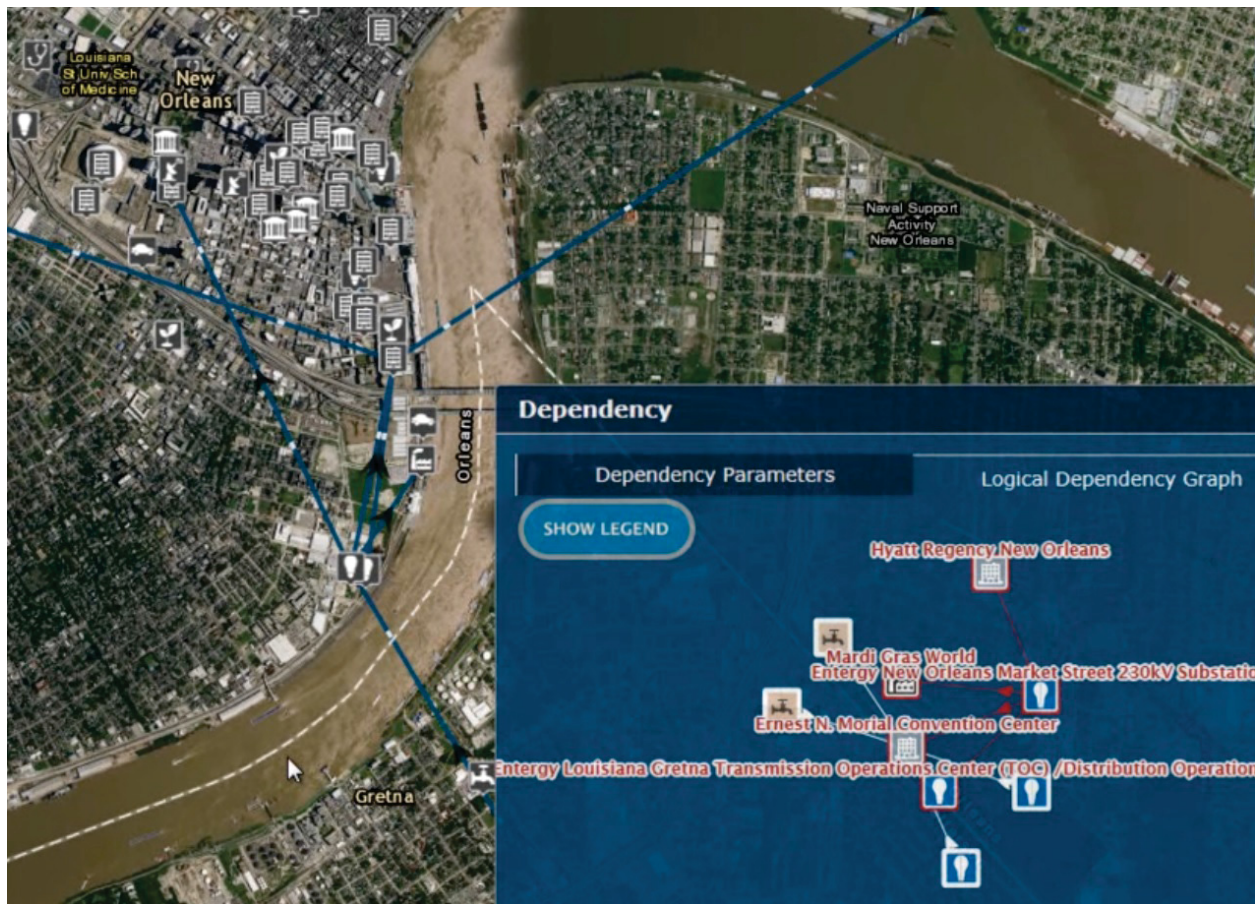


Figure 3. Relationships among CI assets in New Orleans are represented on the map (blue lines with animated white dashes) and on the logical dependency graph (lower right), where the CI assets are located and linked in a statistical and non-geographic position.

## Benefits to INL

Research and development associated with this project has produced tangible and powerful deliverables. These deliverables advance both the technical capabilities of INL, which can be leveraged to secure continued and additional direct-funded projects, and the theoretical underpinnings of geovisual analytics, which will further establish the geospatial core capability of INL's Homeland Security Division as innovators in CI and geo-information research. With these new tools, we are enabling time-critical analysis to enhance resilience of the nation's CI. The new capability represents a major step forward in competencies and business development potential for our division and for other domains within INL that require visualization of large, multi-dimensional, and complex data sets.

## References

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Robinson, A. C., D. J. Peuquet, S. Pezanowski, F. A. Hardisty, and B. S. Swedberg, 2016, "Design and evaluation of a geovisual analytics system for uncovering patterns in spatio-temporal event data," *Cartography and Geographic Information Science*, <http://dx.doi.org/10.1080/15230406.2016.1139467>.

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Edsall, R., 2016, "Interface Modalities," in: *International Encyclopedia of Geography*, Hoboken: John Wiley & Sons (in press; late 2016).

Edsall, R., R. Hruska, S. Cherry, and others, 2016, "Operational and Strategic Solutions for Critical Infrastructure Visualization," *Applied Geography*, in preparation.

## **Presentations**

Cherry, S., 2015, "Visualizing Geospatial Data to Support Critical Infrastructure Interdependency Analysis and Situational Awareness," *2nd National Symposium on Resilient Critical Infrastructure*, Philadelphia, Pennsylvania, August 18 through 20, 2015.

Edsall, R., 2016, "Operational and Strategic Solutions for Critical Infrastructure Visualization," *Annual Meeting of the American Association of Geographers*, San Francisco, California, March 28 through April 1, 2016.

Edsall, R., K. McGillivray, S. Cherry, and R. Hruska, 2016, "Strategic Solutions for High-Density and Interdependency Visualization," *3rd National Symposium on Resilient Critical Infrastructure*, Naperville, Illinois, August 18, 2016.

## **Interns and Postdocs**

Intern: Harvey Hembree

## 15-096—End-to-End Dynamic Program Analysis for Industrial Control Systems with Concolic Execution

Craig Miles,<sup>1</sup> Craig Rieger,<sup>1</sup> and Arun Lakhotia<sup>2</sup>

### General Project Description

Industrial control system (ICS) software drives the nation's critical infrastructure; therefore, every effort must be undertaken to ensure it is free of bugs and exploitable vulnerabilities. Concolic execution is a state-of-the-art program analysis technique that provides for discovery of such bugs and exploitable vulnerabilities in software, making its application to ICS software desirable. However, at its present level of development, the concolic execution technique can only analyze the software running on a single device. In contrast, ICSs are generally comprise multiple devices networked together, with each device running its own software. The software running on the ICS's various devices collectively comprises its software system and would ideally be analyzed as a whole, because bugs and vulnerabilities might only manifest when the devices are working together in concert. The purpose of this research is to extend concolic execution so it can be applied to the entire software system of an ICS. This will further our capability to discover bugs and vulnerabilities in ICSs, thereby, helping ensure their safe and secure operation.

### Summary

Concolic execution serves to discover bugs and vulnerabilities by automatically and repeatedly running a program under many different input values. The goal is to supply a wide enough variety of input values to induce the program into exhibiting many or all of its potential behaviors, including crashes due to bugs and vulnerabilities. The particular input values supplied to the program by concolic execution are generated by analyzing the program's internal logic. When observing the conditions that must be held at the program's decision points, concolic execution can compute input values that satisfy those conditions and drive execution of the program in the desired manner (for instance, determining how we can generate input values derived from observations of the internal logic of programs running on two or more networked devices such as those shown in Figure 1).

The first fiscal year of the project has centered on determining the gaps that must be filled in order to evolve a concolic execution engine such that it can be applied in the ICS domain and also on how those gaps should be filled. For instance, if a programmer incorrectly established a setup that would allow a tank overview in the Figure 1 system, the concolic engine must simultaneously be able to observe execution on two controller devices. Therefore, the first gap has software implementation of the controller, where this execution can be observed. Second, the current state-of-the-art concolic engine does not address floating point operations, which is a limitation when used in evaluating ICS where these operations are prevalent. In addition, the differentiating capability that this research effort would provide is the second gap to be addressed.

During the second year's effort, which concluded after the first quarter, several necessary next steps were taken in collaboration with university partners. A software-based programmable logic controller (PLC) (i.e., an emulator of a four-tank system<sup>3</sup> and human machine interface) were developed as an ICS network to support concolic evaluations.<sup>4</sup> This development is extensible for other use, in addition to providing the underlying framework that allows concolic evaluation. For the latter, access to machine level execution is required and unobtainable from proprietary hardware implementations of a PLC. The other use referred includes a generic PLC and human

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<sup>1</sup> Idaho National Laboratory

<sup>2</sup> University of Louisiana at Lafayette

<sup>3</sup> <http://rcschallenge.inl.gov/FourTankSystem.aspx>

<sup>4</sup> Idaho State University

machine interface implementation for virtualized networks. In particular, this includes showing network traffic with realism within this virtualized environment from sensor data and commands being executed between the human machine interface and PLC.

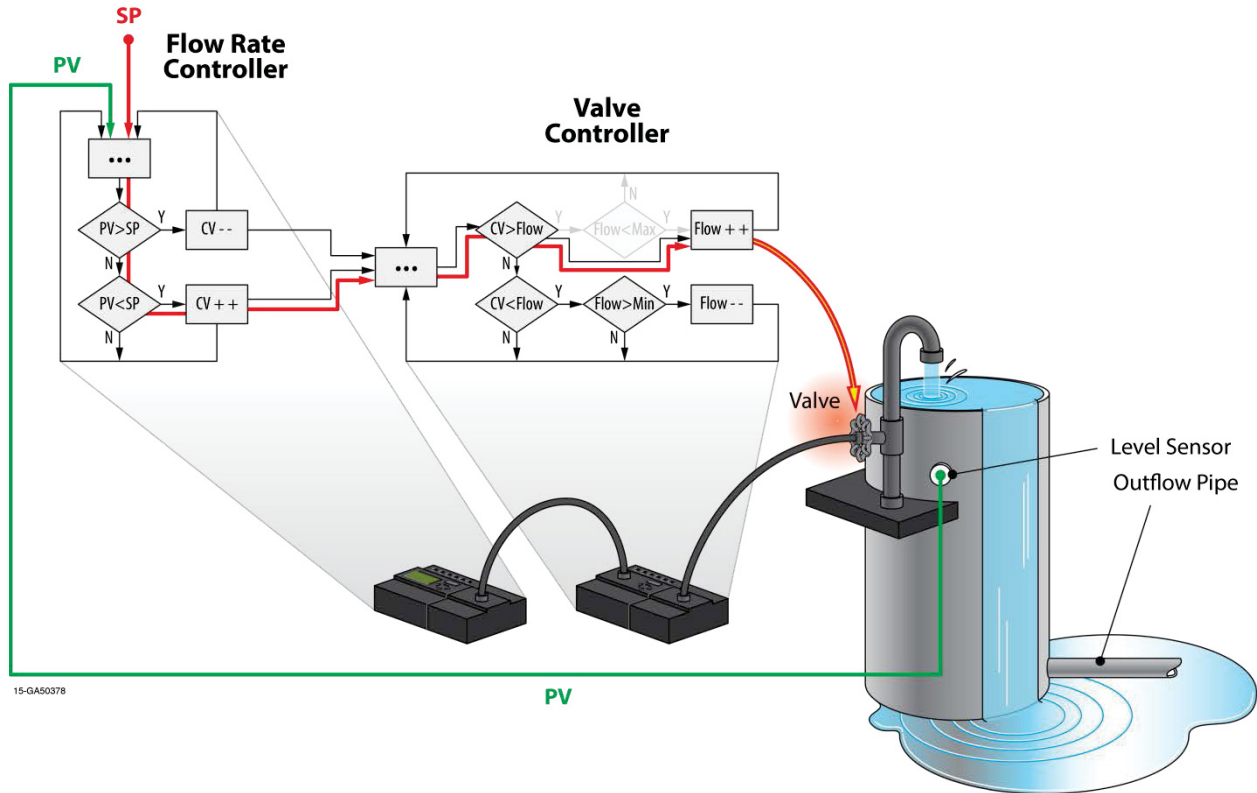


Figure 1. A buggy control system comprise multiple networked devices. Concolic execution for industrial control systems could discover the bug by generating an input value satisfying the decision point conditions along the indicated red path through the logic of both devices' programs.

Further efforts were done to evaluate the extension of floating point operations into the concolic engine used by this effort. This is done to improve the accuracy and remove the dependencies of approximations currently used. Specific recommendations based on current developments from the research and develop community were made, which can be leveraged with limited development to extend the current concolic engine.

## Benefits to DOE

This work will help ensure the cyber security of critical energy infrastructure. In particular, this work will directly meet an objective of DOE's Roadmap to Achieve Energy Deliver Systems Cybersecurity, viz., to Develop and Implement New Protective Measures to Reduce Risk. It advances the state-of-the-art by more effectively recognizing vulnerabilities in the software prior to release.

## Publications

Miles, C., *PowerSPIN Workshop*, Idaho Falls, Idaho, November 10, 2015.

## Collaborations

University: University of Louisiana at Lafayette

### 15-100—Real-Time Process Simulator

Jared Verba<sup>1</sup> and Gordon Rueff<sup>1</sup>

#### General Project Description

The purpose of the Real-Time Process Simulator (RTPS) project is to generate a proof-of-concept simulation framework for industrial control systems (ICS). This research is creating a more open and robust ICS remote input/output simulation framework for devices outside of the Information Technology network. This will enable current and future research efforts to produce more holistic and innovative cyber security solutions for ICS. By developing a field input/output simulation framework that requires hardware-in-the-loop for logic, researchers can look at solutions for protecting a system at the field device level (e.g., the programmable logic controller or remote terminal unit level). This research will yield better understanding and simulation of ICS proprietary protocols, devices, and sensors.

#### Summary

Capability for RTPS has expanded with new research into making the software more user friendly and more accurate than before. RTPS is broken into two different components: (1) a flexible simulation engine and (2) a network protocol translator.

The simulation portion is an expandable system that can take multiple sources, including feeds from an INL Multi-Physics Object-Oriented Simulation Environment (known as MOOSE) model and generate signals for a process control system. This is beneficial if a user wants to use high-fidelity models for select components of the simulation, but does not want to spend time or the computational resources of having the whole simulation run at high fidelity.

The protocol translator is capable of receiving multiple data feeds and producing a protocol that can be understood by a Siemens programmable logic controller. The data feeds use a common protocol that can be adapted to various outputs, allowing multiple computers and simulation systems to feed their output into the translator to produce a single image of a nuclear system or ICS. This research is unique because it uses current ICS hardware for process control and can be run with minimal resources. The end goal of this research is to create a proof-of-concept with direct applicability to the growing nuclear cyber business, current ICS training, and future security tools for ICS networks. Figure 1 shows how RTPS can be used to simulate a real work system. By using RTPS coupled to hardware-in-the-loop and existing nuclear simulations, a realistic and safe simulation of nuclear facilities can be achieved. Future work on RTPS will include building a simulation of a current nuclear centric real-world process housed at INL.

#### Benefits to DOE

This research is directly applicable to the current and ongoing need for security in the ICS and nuclear fields. This project is aligning with the goals of the upcoming Cybercore Innovation Center to provide a unique capability for DOE and INL. This project also meets a need for better research tools on ICS and nuclear systems.

#### Publications

A whitepaper describing the concept and benefits of the research has been provided to current customers looking for an alternative to large testing facilities.

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<sup>1</sup> Idaho National Laboratory

Presentations are given to current customers seeking a method of using simulation in large-scale training to save on cost and maintenance of large physical facilities and ICS processes.

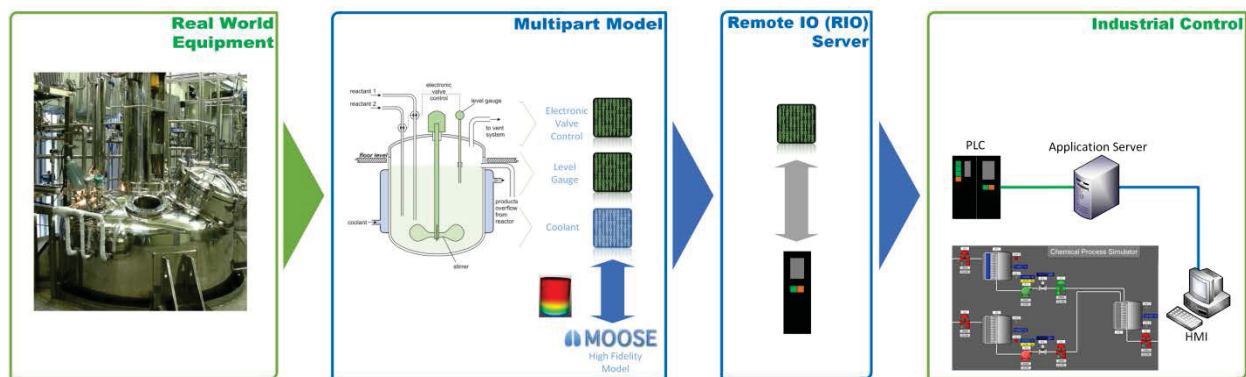


Figure 1. RTPS applied to a real-world process.

## Invention Disclosures, Patents, Copyrights

An IDR was filed for this project this year. It is currently in the processing phase.

## Interns and Postdocs

Interns: Naomi Hazelip and Keith Drew

## 15-111—Adversary Signature Development and Threat Analysis

Sarah Freeman<sup>1</sup> and Katya Le Blanc<sup>1</sup>

### General Description

Cyber security is most effective before an attack. However, the development of adequate proactive protection remains a challenge. Recent years have seen an explosion of threat-focused research aiming to meet the needs of an ever-evolving cyber landscape. Organizations, both public and private, have sought to advance threat analysis processes, methodologies, and capabilities in order to improve capabilities and training for the next generation of defenders. This research will improve our understanding of the cyber community by developing additional structured analytical techniques and a corresponding model to address the current challenges facing complex threat analysis.

### Summary

This research blends the current concepts of behavioral economics with the analytical procedures used in law enforcement and the intelligence community in order to fill the current gap in threat analysis capabilities. The goal of this work is greater understanding of how hackers interact, share information, approach problems, and use technology. Central to this process is the development of a model that more fully describes attack scenarios and campaigns. The model is supported by a collection of indicators (such as exploits and malware used), which serve as the basis for attribution. The research conducted during FY 2015 covered two related, but uniquely focused, phases for the identification, development, and verification of a new collection of indicators. Data collected from these two phases has been combined into a single model, which will serve as a library of information for the development of cyber adversary profiles (see Figure 1).

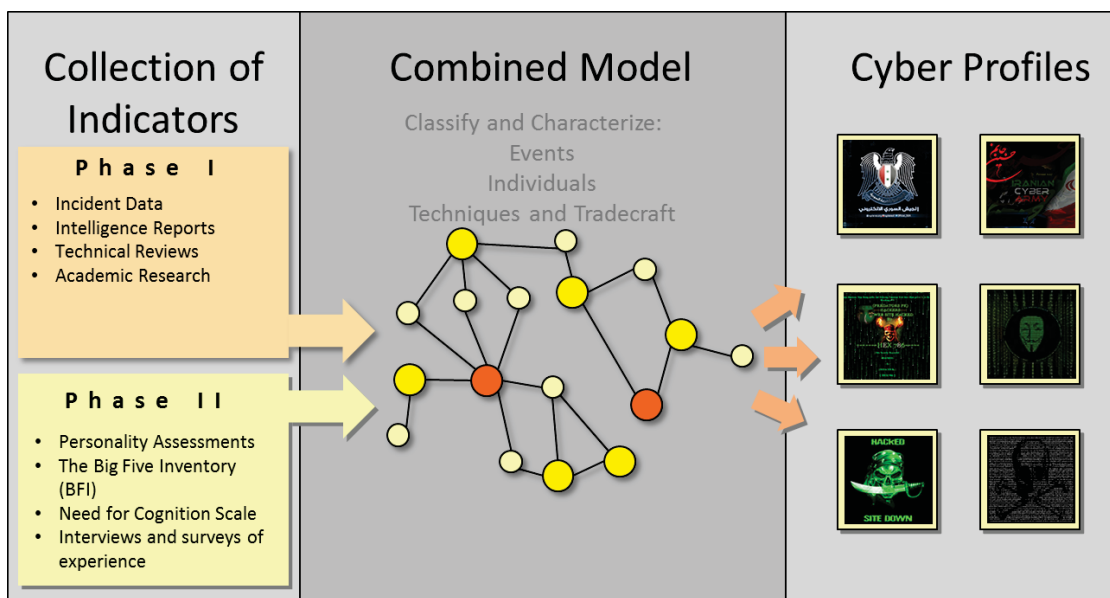


Figure 1. The experimental design was constructed to include both objective and subjective indicators for ontology development. This model then serves as the analytic framework for the creation of cyber profiles.

<sup>1</sup> Idaho National Laboratory



A critical component of Phase I included development and analysis of cyber incidents. The goal of this process was two-fold. First, it ensured a wider perspective was considered for both this research and the development of the combined model, because it was assumed that the cyber researcher participants were not representative of the complete hacker culture. Second, post-mortem analysis of cyber incidents assisted with the aggregation of ‘typical’ indicators (i.e., the components most often tracked and recorded following a cyber incident). During the second year of research, this work incorporated input from researchers from the Graduate School of Public and International Affairs at the University of Pittsburgh.

The experimental design for Phase II included two vetted psychological assessment tools (i.e., the Need for Cognition Scale and the Big Five Inventory), as well as an INL-designed structured interview protocol to investigate the experiences, skills, and technology preferences of the cyber security community. By expanding quantitative data beyond that traditionally collected post-incident, the research team was able to develop a more comprehensive picture of the personal motivations and thought processes of these individuals. By the same token, the inclusion of cyber incident data allowed the research team to evolve the ontology further than would be possible with the interviews alone, especially given the size of the potential sample pool. The end result of this holistic approach is a more comprehensive picture of potential signature components that can be expanded in the future.

## Benefits to DOE

This research contributes directly to DOE’s goal of securing critical infrastructure from cyber attacks by evaluating the existing cyber researcher community’s methods and capabilities. This novel threat analysis approach will allow organizations to more accurately characterize the modern cyber threat actor, promoting creation of evolved industrial control systems protection systems. Additionally, findings from this work will allow us to more fully define the necessary skills for developing the next generation of cyber defenders.

## Publications

Le Blanc, K. and S. Freeman, 2016, “Investigating the Relationship between Need for Cognition and Skill in Ethical Hackers,” INL/CON-15-37384, publication in *Advances in Intelligence Systems and Computing*, proceedings of the *7th International Conference on Applied Human Factors and Ergonomics*, Orlando, Florida, July 2016.

## Presentations

Freeman, S., 2015, “Expanding the Cyber Portfolio,” *RCTCon2015*, Baltimore, Maryland, December 9 and 10, 2015.

Freeman, S., 2015, “Adversary Signature Development and Threat Analysis,” *Strategic Advisory Committee Poster Session*, Idaho National Laboratory, June 25, 2015.

Freeman, S., 2015, “Expanding the Cyber Portfolio,” *Early Career Researcher Symposium*, Idaho National Laboratory, July 30, 2015.

Freeman, S., 2016, “Adversary Signature Development and Threat Analysis,” *Strategic Advisory Committee Poster Session*, Idaho National Laboratory, June 30, 2016.

Le Blanc, K. and S. Freeman, 2016, “Investigating the Relationship between Need for Cognition and Skill in Ethical Hackers,” *7th International Conference on Applied Human Factors and Ergonomics*, Orlando, Florida, July 29, 2016.

## **Interns and Postdocs**

Interns: Rachael Hill, Andrew McClusky, Cynthia Chavez, Brendan Lyle, Kevin Pollock, Abby Waliga, Michael Goldstein, and Alex Lin

## 16-081—Modeling Thermite Reactions

Ron Heaps,<sup>1</sup> Bryon Curnutt,<sup>1</sup> Hai Huang,<sup>1</sup> Jing Zhou,<sup>1</sup> Ron Heaps,<sup>1</sup> Dr. Pantoya,<sup>2</sup> Ryan Bratton,<sup>2</sup> and Jason Green<sup>2</sup>

### General Project Description

This effort focuses on developing a model that emulates thermite reactions using the Multi-Physics Object-Oriented Simulation Environment (MOOSE) framework. The finite-element multi-physics framework MOOSE is being used to provide a coupled physics simulation tool to model these highly exothermic reactions for the benefit of INL and the scientific community.

### Summary

The effort this year has been to port work done by C. R. Bowen and B. Derby, “Finite-Difference Modelling of Self-Propagating High-Temperature Synthesis of Materials,” 1994. The reaction cited follows the chemical composition  $3\text{TiO}_2 + 4\text{Al} + 3\text{C} \rightarrow 3\text{TiC} + 2\text{Al}_2\text{O}_3 + \text{Energy}$ . It was decided that this paper would provide a good starting point to set up in MOOSE. Bowens work is based on a finite difference and has the following benefits. It is a model set up as a one-dimensional mesh, it is a solid-solid reaction, phase changes are not accounted for, and it is a base to develop and check our model. We have been able to build a similar model under the MOOSE framework. The model deals with propagation of the reaction and tracks the reaction wave in the material compact. More complexity will be added as the model matures. For example, elements will be added to account for the latent heat required for the phase changes of the reactants. At this point, our MOOSE application will be referred to as Raptor. Raptor is built on a two-dimensional mesh (10 x 100). The current model version generates an output file that tracks the reaction well and is beginning to follow what is seen in the experimental data and publications.

Heat transfer is the primary driver for the combustion wave in these exothermic reactions. The heat transfer modules, specifically the heat conduction and heat conduction time derivative kernels, are readily available in the MOOSE framework. Steady-state heat conduction is given by:

$$-\rho C_p \frac{\delta T}{\delta t} + \nabla \cdot (-k \nabla T) - Q = 0$$

The weak form of the equation is then input into MOOSE syntax. Similarly, heat convection, radiation heat transfer, and a number of other applicable heat transfer boundary conditions already utilized by Raptor are available in the MOOSE framework. The current approach used in Raptor is to quickly input  $T_{\text{ad}}$  and the ignition temperature ( $T_{\text{ig}}$ ) of the reaction, or the temperature at which the exothermic reaction takes place, to the Raptor input file rather than being calculated by Raptor. These values are then passed to Raptor’s forcing function. An additional materials kernel has been added to assign the material properties for both the products and reactants.

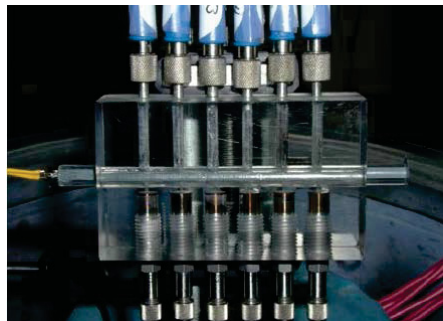
Texas Tech has been working on experiments that will be used to verify Raptor. Ryan Bratton has been working to determine the reaction rate experimentally using a Bockmon tube setup. The Bockmon tube is an acrylic tube that is filled with powder and reacted (see Figure 1). The tube can also be instrumented to gather data such as pressure and temperature, as well as viewing the reaction.

Light is emitted during the reaction and has usually been used to track the reaction front but can lead to oversaturation of the camera sensor (see Figure 2) Ryan has been developing a system that illuminates the reaction using a laser. The combination of laser illumination, high speed camera, and filters, captures the reaction

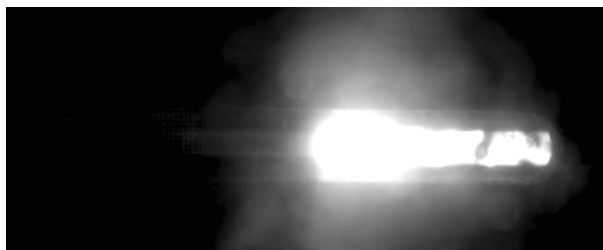
<sup>1</sup> Idaho National Laboratory

<sup>2</sup> Texas Tech University, Texas

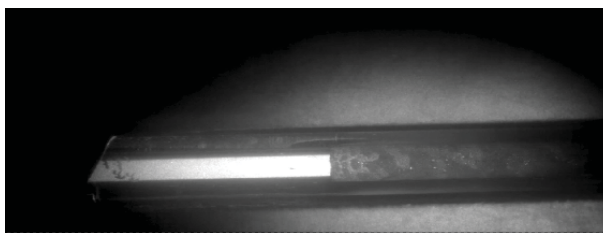
front in great detail (see Figure 3). More accurate measurements can be made when the reaction front is well defined. In addition to better resolution, this system tends to flip the bright and dark portions of the image. This can be seen in Figures 2 and 3. The white part of Figure 2 is self-illumination due to the reaction; the dark area to the left is the unreacted compact. Figure 3 uses a laser to illuminate the experiment. The white part is the reflection of the unreacted compact and the darker area to the right of the reaction front is the reaction. Experimental data collected from these and other experiments will be used in development of our model.



*Figure 1. Bockmon tube setup.*



*Figure 2. Self-illuminated.*



*Figure 3. Laser illuminated.*

Much work still needs to be done to expand Raptor to the point it is useful in research. Once established, it will allow initial research to be done more cost effectively and provide increased safety by being able to work in a simulation environment.

### **Benefits to DOE**

Reactions of this type are used in many devices made for DOE missions. They have been used to test a nuclear reactor vessel's ability to withstand meltdown conditions. Devices are made and used within national security to test DOE infrastructure. There have been little modeling efforts for thermites. This project will also be a benefit to basic science. In addition, related developments (e.g., illumination system) have the potential be used in other DOE projects.

## **Interns and Postdocs**

Intern: Jason Green and Ryan Bratton

Postdoc: Jing Zhou

## **Collaborations**

University: Texas Tech University

## 16-106—ICS-CAPE: Industrial Control Systems – Cyber Attacks and their Physical Effects

Mary Klett,<sup>1</sup> Dale Christiansen,<sup>1</sup> and Tim Klett<sup>1</sup>

### General Project Description

Industrial control systems (ICS) cyber-attacks and their physical effects (CAPE) is focused on expansion and improvement of the nation's capability to identify and understand the physical impacts a cyber-attack on an ICS may have and categorize data points at both a general and industry-specific level in order to identify functional consequences.

Diving into the inner workings of a component/system/asset will enable a robust assessment of a specific facility or collection of industry assets. This assessment will lead to a more accurate understanding of what the potential functional consequence of a cyber-attack on an ICS network may be. Building the component profiles on an established platform will allow analysis to be conducted within a facility and taken one level out to identify potential impacts to supported infrastructure resulting in a cumulative consequence.

Many models and methodologies used to determine the best way to protect or attack an ICS network exist; these approaches oftentimes consider the worst-case scenario, but, in actuality, there is a gap in methods for aiding in understanding the physical effects of a cyber-attack. The goal of this research is to architect a base structure that can be used to develop a library of infrastructure-specific process maps and identify key data points to be collected at each connection or component in order to determine functional impacts.

This research will assess and enhance the applicability of using a universal model to delve into components typically used within an industry for enabling detailed evaluation of the impact of an attack or failure of a specific asset. The universal model will allow for rapid analysis of data points both across and within specific industries. It will identify the concrete consequences of a cyber-attack and enhance understanding of the potential for cascading impacts of a cyber-attack on a specific component or asset.

### Summary

A high-level catalog of asset types and expected complexity was developed based on the Department of Homeland Security's infrastructure taxonomy. In turn, this will be used to build out mapping profiles for asset types.

Electric substations were selected as the first candidate for development of a profile. A thorough literature review was conducted and an initial profile was developed. Subject matter expert (SME) interviews were conducted in order to review the information collected and identify any additional critical points. After SME interviews, it was determined that the overall function and characteristics for these components were similar enough to allow for grouping by component type; therefore, paring the list down from more than 50 to just over 10. After the initial interviews, we regrouped and developed templates that were sent back to the SMEs for review and approval.

In addition to building out mapping profiles at an asset-type level, specific facility or asset base data points that will be collected at each component were identified. These include questions regarding the following:

- Criticality to operations or support to critical components
- Any redundancies that exist and details on that redundancy
- Any safeguards or countermeasures and relevant details that exist.

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<sup>1</sup> Idaho National Laboratory



Additional data points may be collected based on the industry and/or component type. However, the base data points that will be used to determine initial functional consequence are those listed above.

Initial review and interviews for profiling water treatment plants have begun.

## **Benefits to DOE**

Research undertaken in this project supplements DOE's role as the sector-specific agency and national leader in energy/electric grid security against cyber-attacks. The resulting methodology stemming from this work will provide benefit to Department of Homeland Security personnel, including protective security advisors and cyber security advisors, so the physical impacts and resulting cascading consequences of a cyber-attack can be better understood. These Department of Homeland Security advisors are engaged with critical infrastructure owners and operators, and this work will supplement Department of Homeland Security efforts to secure the electric grid and other critical infrastructure types.

## **Publications**

### ***Presentations***

"Transforming the Resilience of Cognitive Cyber-physical Systems," Lightning Round Presentation, *Resilience Week 2016*, Chicago, Illinois, August 16 through 18, 2016.

## **Interns and Postdocs**

Interns: Karen Kursteiner

Postdoc: Gabriel Weaver

## **Collaborations**

University: Carnegie Mellon

## 16-133—Secure SCADA Communications System

Briam Johnson,<sup>1</sup> Kris Watts,<sup>1</sup> and Stephen Kleinheide<sup>1</sup>

### General Project Description

This research is based on the hypothesis that industrial control system (ICS) endpoints, including the software that monitors and controls the system, controllers, and intelligent devices are all vulnerable to cyber-attack. Many of these devices remain in service for 20 years or more; therefore, even if secure solutions were available today, it would take decades to solve this problem. Instead of trying to solve this very large problem, our approach is to add capability to the portion of the system that connects these devices—the communications system. In the majority of cases, network switches or similar devices provide the infrastructure that connects ICS components. These devices are much easier and less expensive to replace than the control system endpoints.

The primary benefits of the system we are trying to develop include enhanced situational awareness, out-of-band operator control of network flows, and real-time flagging of exceptions.

### Summary

During this first year of the project, we began by integrating a simple electric sector-focused control system consisting of a single human machine interface computer and two remote terminal units from different manufacturers. We chose a commonly used, open protocol as an example for initial development. Note that this technology is designed to be vendor agnostic, but does require a protocol-specific module for every protocol used in a system. Fortunately, most modern systems use a relatively small handful of open protocols.

In lieu of a traditional Ethernet switch, we implemented a software-defined network switch using open source software for basic functions. We created custom software to implement specific rule sets to prevent and flag unauthorized messages.

To allow for operator monitoring and control, we built a basic graphical user interface that allows the user to view the network flows in real-time, enable or disable control on a per-device or system-level (i.e., panic button) basis, and see a list of exceptions that occur.

During course development, we learned several things that have made this research more challenging. One issue is that not every device that uses the same protocol responds the same to a given message. In some cases, the slightest interruption in communications causes the device to lock up in a way that can only be fixed with a hard reboot. In general, the control equipment is very intolerant to any disruption in communications and/or any unexpected message sequences. This makes implementation of our system more difficult, but underscores just how vulnerable these devices are to cyber-attack. Nevertheless, we have found ways to work around the above issues and have met our primary Year 1 objective of developing a proof-of-concept demonstration, showing that is possible to create a system as described above.

### Benefits to DOE

This research directly supports INL's mission to discover, demonstrate, and secure innovative nuclear energy solutions, other clean energy options, and critical infrastructure. Specifically, it is aimed at trying to answer several key questions with respect to securing ICSs that monitor and control critical infrastructure:

1. How can we detect when attackers are probing ICS networks?
2. How do we prevent attacks from propagating once an attacker gains access?

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<sup>1</sup> Idaho National Laboratory

### 3. How do we limit or eliminate an attacker's ability to cause damage during an ongoing attack?

The recent attack on Ukraine's electrical infrastructure demonstrated these needs. Other work is ongoing to answer some of these questions; however, the last two questions, in particular, are ones that we believe this research can help answer where many other solutions stop short.

It is clear that new, more sophisticated and control-system focused security tools are required to thwart the efforts of ever more sophisticated attacks. If successful, this research could help mitigate immediate cyber threats to critical infrastructure.

### **Interns and Postdocs**

Staff: Stephen Kleinheider

## 16-152—Wireless Radiofrequency Signal Identification and Protocol Reverse Engineering

Kurt Derr<sup>1</sup> and Samuel Ramirez<sup>1</sup>

### General Project Description

Several prominent wireless communications protocols (e.g., WiFi, Bluetooth, ZigBee, and LTE) are in use today in addition to proprietary protocols. Wireless technology is proliferating with the advent of low-cost computers and devices for the internet of things. The expanding role of wireless communications has created a need for rapid identification of wireless protocols in use by some black box devices (i.e., an arbitrary device that arrived in the mail that has a radio and uses proprietary/ unknown protocols) or wireless computing systems (i.e., a computer system that uses a new proprietary protocol).

The objective of this project is to build a tool, Wireless radioFrequency signal Identification and Protocol Reverse Engineering (WiFIRE), usable in an operational context for automatically analyzing the radiofrequency emanations and wireless protocols from a black box device and performing automated wireless protocol reverse engineering. The hypothesis is that GNU Radio's (i.e., a software-defined radio) computational intelligence techniques (e.g., neural networks) and open source protocol reverse engineering tools may be combined to perform radio signal detection, classification, demodulation, and reverse engineering of network protocols.

WiFIRE will scan multiple frequency ranges looking for wireless signals of interest emanating from a black box. The goal is to infer protocols from these signals and reverse engineer the protocols so that penetration testing may generate network protocol inputs to the black box wireless protocol, uncover and exploit potential vulnerabilities, and gain control of the device, which is a novel capability.

### Summary

During the first year of the project (FY 2016), multiple spectrum sensing algorithms were implemented that were used to identify and classify wireless communications traffic. Spectrum sensing is the task of obtaining awareness about spectrum usage in a geographical area or of a specific device in a radiofrequency-isolated environment. These spectrum sensing algorithms will undergo further enhancement to broaden the wireless classification capabilities that are a major component of the WiFIRE tool under development.

Classifying wireless communications traffic requires knowledge of the signatures of specific types of wireless signals such as WiFi, Bluetooth, and ZigBee. A test environment was created that enabled the capture of specific wireless signals in the absence of interference from other wireless activities. These results were reported in a publication developed, submitted, and accepted for the GNU Radio Conference in September 2016.

This important tool under development allows INL researchers to view all ongoing wireless communications activities to identify both expected and unexpected wireless communications from a device or in a geographical area. Although many other organizations have the capability to view wireless signals at a specific frequency range, INL is one of few that may examine wireless signals across the spectrum, classify the signal, and reverse engineer the protocols in use. This tool could prove to be a very important milestone for further development of new tools that improve wireless security and mitigate system threats.

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<sup>1</sup> Idaho National Laboratory

## Benefits to DOE

This capability will enable INL/others to determine the network protocols in use by a wireless black box device or in a geographical area that may pose a threat to the government and INL customers. WiFIRE will also enable the potential discovery of exploitable black box device vulnerabilities that may lead to gaining control of the device.

Software-defined radio is dramatically accelerating the pace of innovation and development of new radio technologies and enabling development of tools that were previously not possible without unique hardware and software. Wireless communications devices are widely used in public safety, emergency response, and critical infrastructure applications. WiFIRE will enable the government to identify the wireless protocols used by a black box device, assess the robustness of wireless implementation, analyze traffic, identify potential data leakage, and compare implementation of a protocol with its official specifications.

The novelty of WiFIRE is that no real tool is currently available for capturing radiofrequency data, quickly identifying the protocols in use, and performing protocol reverse engineering.

## Publications

Hearn, C., A. Kuznicki, C. Becker, K. Derr, and S. Ramirez, "Data Acquisition in Wireless Router Link Testbed using GNU Radio Companion," *GNU Radio Conference (GRCON 2016)*, September 12 through 16, 2016, accepted.

## Interns and Postdocs

Interns: Christopher Becker, Aniqua Baset, and Andrew Kuznicki

## Collaborations

University: University of Utah

## **16P6-007FP—Consequence-Driven Cyber-Informed Engineering Methodology Pilot**

Robert Smith,<sup>1</sup> Sarah Freeman,<sup>1</sup> Colleen Glenn,<sup>1</sup> and Curtis St. Michel<sup>1</sup>

### **General Project Description**

This project team is engaging in a high-impact effort to prioritize high-consequence cyber risk to industrial control systems of the country's most critical infrastructure. This pilot project will include building a methodology to inform the conduct of an in-depth engineering evaluation of a major electric utility's operational environment in order to identify and mitigate high-consequence cyber attack scenarios. This consequence-based risk analysis methodology of the integrated control system environment will be leveraged by the team to propose new engineering or design options, operational procedures, and active defense methods, alerts, and/or safeguards. As part of this process, the team will build a method for how to work with the intelligence community to baseline current adversarial capabilities and identify whether these skills could be applied to attack an electric utility's operational processes.

### **Summary**

The project was initiated on September 1, 2016; therefore, we have just begun substantive work. As we get further into the project, we plan to use the results to validate a consequence-driven, cyber-informed engineering (CCE) framework and identify key lessons learned that can be applied to the broader energy sector. The general, long-term goal of this project is to develop and hone the CCE process so any organization can use CCE to evaluate and improve their cyber security posture autonomously, regardless of size or scale.

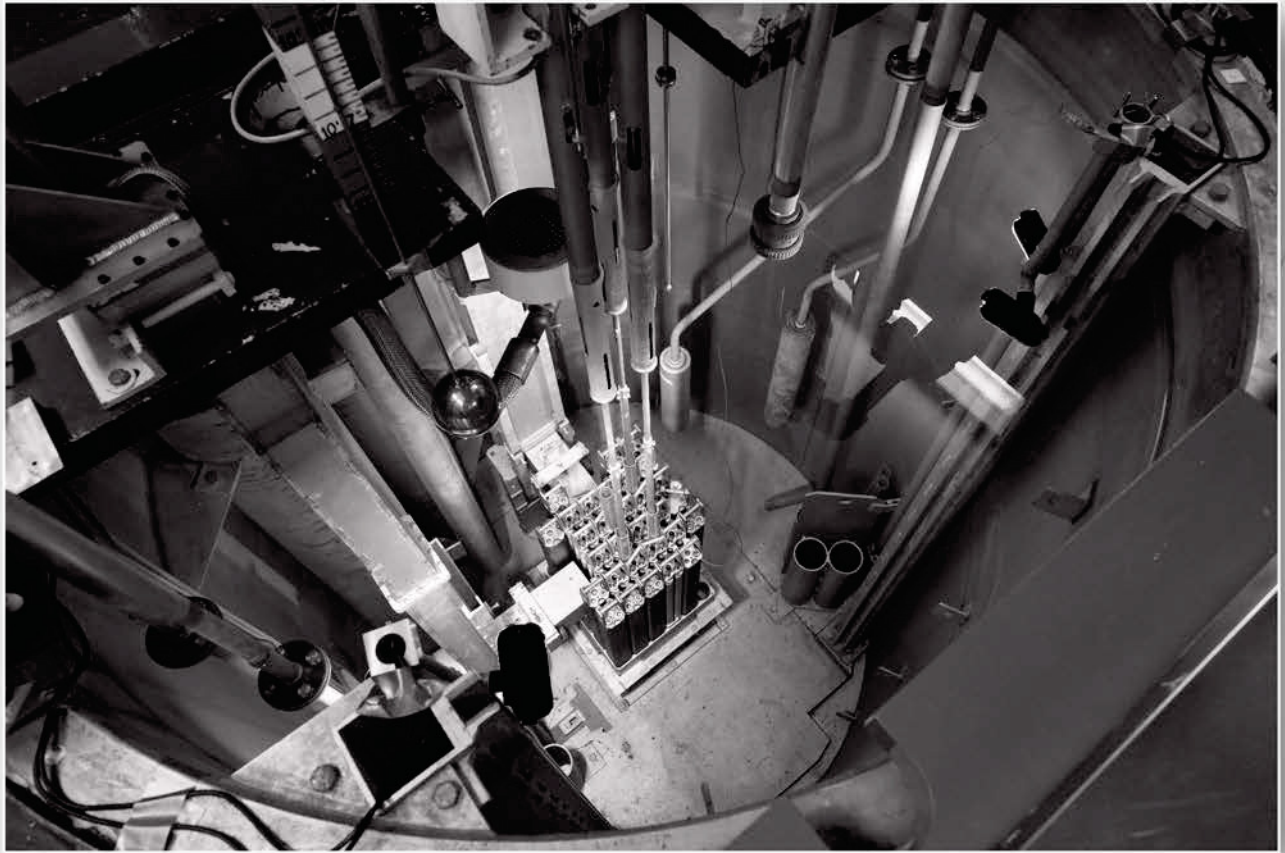
### **Benefits to DOE**

The team believes that this process, once demonstrated and validated within the electric sector, can be applied to the oil and gas industries and to military and government assets. As we get further into the work of the project and this initiative succeeds, INL and the U.S. government will be able to continue their leadership role in providing industry with practical, innovative solutions for securing the nation's critical infrastructure.

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<sup>1</sup> Idaho National Laboratory





# Appendices



## **Appendix A**

### **Benefiting Agencies**



## Appendix A

### Benefiting Agencies

The following table provides a listing of individual LDRD project contributions to each benefiting agency. Green dot represents the primary beneficiary and the black dots represent secondary beneficiaries. In addition, FY 2016 total investments are provided for each benefiting agency.

Project Number	PI Name	Title	Department of Homeland Security	Department of Defense	DOE Office of Energy Efficiency and Renewable Energy	DOE Office of Environmental Management	DOE Office of Environmental, Safety, and Health	DOE Office of Fossil Energy	DOE-NE	National Nuclear Security Administration	DOE Office of Naval Reactor	DOE Office of Civilian Radioactive Waste Management	DOE-Office of Electricity Delivery and Energy Reliability	DOE Office of Science
14-009	James E. O'Brien	Development of an INL Capability for High-Temperature Flow, Heat Transfer, and Thermal Energy Storage with Applications in Advanced Small Modular Reactors, High-Temperature Heat Exchangers, Hybrid Energy Systems, and Dynamic Grid Energy Storage Concepts			•				•				•	
14-010	Kurt Davis	Use of Linear Variable Differential Transformer-Based Methods to Detect Real-Time Geometry Changes during Irradiation Testing		•					•		•			

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Project Number	PI Name	Title	Department of Homeland Security	Department of Defense	DOE Office of Energy Efficiency and Renewable Energy	DOE Office of Environmental Management	DOE Office of Environmental, Safety, and Health	DOE Office of Fossil Energy	DOE-NE	National Nuclear Security Administration	DOE Office of Naval Reactor	DOE Office of Civilian Radioactive Waste Management	DOE-Office of Electricity Delivery and Energy Reliability	DOE Office of Science
14-025	Mitchell Greenhalgh	Minor Actinide and Lanthanide Separations in Alternative Media				•			•			•		•
14-026	Yongfeng Zhang	Multi-Scale modeling on Delayed Hydride Cracking in Zirconium: Hydrogen Transport and Hydride Nucleation							•			•		•
14-031	Richard Williamson	Multi-Dimensional Multi-Physics Modeling of Fuel Behavior During Accident Conditions							•					
14-032	Jonathan Chugg	Remote Vulnerability Analysis of CAN Bus Networks	•		•								•	
14-037	David Chichester	Advanced Nuclear Material Characterization Technology for Security Applications	•			•	•		•	•		•		•
14-041	Robert O'Brien	Fabrication of UN-U <sub>3</sub> Si <sub>2</sub> via Spark Plasma Sintering							•					
14-045	Scott J. Thompson	End-to-End Radiation Detector Enhancements for Improved Safety and Security in Safeguarded Facilities	•						•	•				•
14-075	Congjian Wang	Development of Tools and Methodologies for Uncertainty Quantification and Validation for Multi-Physics Fuel Performance Simulation							•					



Project Number	PI Name	Title	Department of Homeland Security	Department of Defense	DOE Office of Energy Efficiency and Renewable Energy	DOE Office of Environmental Management	DOE Office of Environmental, Safety, and Health	DOE Office of Fossil Energy	DOE-NE	National Nuclear Security Administration	DOE Office of Naval Reactors	DOE Office of Civilian Radioactive Waste Management	DOE-Office of Electricity Delivery and Energy Reliability	DOE Office of Science
14-078	Laura Carroll	Extended Stability Gamma-Gamma Prime Containing Nickel-Base Alloys			●			●	●				●	
14-079	Aaron D. Wilson	Second Generation Switchable Polarity Solvent Draw Solutes for Forward Osmosis			●	●	●	●	●					●
14-086	Kurt S. Myers	Development of a Microgrid/Smartgrid Testbed for ESL and Super Lab Initiative with Load Variability characterization and Control for Renewable Energy Integration			●	●		●	●	●			●	
14-093	Ryan C. Hruska	All Hazards Critical Infrastructure Knowledge Framework	●		●								●	
14-094	Henry Chu	SMC Advanced Armor Materials and Systems Research and Development: Scale-Up Demonstration of Next Generation Armor Steel								●			●	●
14-095	Gary S. Groenewold	In Situ Measurement of Electrolyte Chemistry in Battery Cells During Operation			●								●	
14-098	Jian Gan	Irradiation Effects of Uranium Dioxide							●					●
14-104	Richard C. Martineau	Development of a Multi-Physics Algorithm for Analyzing the Integrity of Nuclear Reactor Containment Vessels Subjected to Extreme Thermal and Overpressure-Loading Conditions		●					●		●			●

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Project Number	PI Name	Title	Department of Homeland Security	Department of Defense	DOE Office of Energy Efficiency and Renewable Energy	DOE Office of Environmental Management	DOE Office of Environmental, Safety, and Health	DOE Office of Fossil Energy	DOE-NE	National Nuclear Security Administration	DOE Office of Naval Reactor	DOE Office of Civilian Radioactive Waste Management	DOE-Office of Electricity Delivery and Energy Reliability	DOE Office of Science
14-106	Joshua Kane	Understanding the Growth of Ultra-Long Carbon Nanotubes			●	●	●		●				●	
15-002	Peter Zalupski	Experimental Scenarios of Adversity and Recovery in Aqueous Separations.			●	●			●	●				●
15-013	Curtis Smith	Simulation-Based Analysis of Procedures and Accident Management Guidelines							●					
15-023	Justin Coleman	Development of Stochastic Three-Dimensional Soil Response Capability in MOOSE to Provide Design and Beyond Design Basis Seismic Motions for Nuclear Facilities				●			●	●				
15-032	Krzysztof Gofryk	Development of New Method for High-Temperature Thermal Conductivity Measurements of Nuclear Materials							●					●
15-039	Shannon Bragg-Sitton	Transient Modeling of Integrated Nuclear Energy Systems with Thermal Energy Storage and Component Aging and Preliminary Model Validation via Experiment			●			●	●				●	
15-040	Vivek Agarwal	Acoustic Telemetry Infrastructure for In-Pile ATR and Treat Monitoring			●				●					

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15-060	Mark DeHart	Development of Efficient TREAT Modeling with Graphite Data Improvement							●	●				
15-083	Shane Cherry	Visualizing Highly Dense Geospatial Data	●		●									●
15-094	James Disser	Risk-Informed Safety, Security, and Safeguards by Design Guidance for Advanced Reactor Concepts							●	●				
15-096	Craig Rieger	End-to-End Dynamic Program Analysis for Industrial Control Systems with Concolic Execution	●										●	
15-100	Jared Verba	Real-Time Process Simulator	●		●								●	
15-111	Sarah Freeman	Adversary Signature Development and Threat Analysis	●						●				●	
15-125	Eric Dufek	Phosphoranimines for Advanced Battery Applications			●								●	●
15-128	Hai Huang	Microstructural Evolution and Mesoscale Coupled Flow-Reaction-Fracturing Processes in Organic Rich Nanoporous Shales			●	●		●						●
15-135	Manish Mohanpurkar	Dynamic Simulations for Large-Scale Electric Power Networks in Real-Time Environment using Multiple RTDS			●				●				●	●

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Project Number	PI Name	Title	Department of Homeland Security	Department of Defense	DOE Office of Energy Efficiency and Renewable Energy	DOE Office of Environmental Management	DOE Office of Environmental, Safety, and Health	DOE Office of Fossil Energy	DOE-NE	National Nuclear Security Administration	DOE Office of Naval Reactor	DOE Office of Civilian Radioactive Waste Management	DOE-Office of Electricity Delivery and Energy Reliability	DOE Office of Science
15-140	Kevin Gering	Expanding the Utility of Advanced Chemical Physics Models for Electrolytes			●								●	●
15-141	Hongbin Zhang	Interfacing MOOSE Components to Enhance Capability							●	●				
15-142	Sebastian Teyssyre	New In-Core Neutron Diagnostics							●	●				●
15-144	Shelly Li	Investigation of Sonication-Assisted Electrolytic Reduction of Used Oxide Fuel in Molten Salt				●			●					
15-145	James Keith Jewell	Advanced Neutron and X-Ray Imaging at TREAT		●					●	●	●		●	●
15-146	Rebecca Fushimi	Tailoring the Kinetic Function of a Surface through Electronic Effects of Nanoscale Architecture			●			●						●
16-002	Chenlin Li	Advanced Carbon Feedstock Processing Using Ionic Liquids			●	●		●						●
16-003	Prabhat Tripath	Recycling of Tantalum-Containing Waste Materials to Recover Tantalum Metal	●		●	●			●					●
16-009	Troy Unruh	Change Detection System for Nuclear Applications	●	●	●			●	●		●		●	

Project Number	PI Name	Title	Department of Homeland Security	Department of Defense	DOE Office of Energy Efficiency and Renewable Energy	DOE Office of Environmental Management	DOE Office of Environmental, Safety, and Health	DOE Office of Fossil Energy	DOE-NE	National Nuclear Security Administration	DOE Office of Naval Reactor	DOE Office of Civilian Radioactive Waste Management	DOE-Office of Electricity Delivery and Energy Reliability	DOE Office of Science
16-010	Daniel Schwen	Development of a Fully Coupled Radiation Damage Production and Evolution Simulation Capability							●	●				
16-013	Suleyman B. Biner	Micromechanistic Approach and Critical Experiments for Quantitative Predictions of Delayed Hydride Cracking in Zirconium Alloys							●					●
16-017	Haihua Zhao	Evaluation of Load-Following Capabilities of Existing and New Nuclear Power Reactors in the Grid with Large-Scale Renewable Energy Sources			●				●				●	
16-026	Robert Youngblood III	Computationally Efficient Prediction of Containment Thermal Hydraulics Using Multi-Scale Simulation: Feasibility Study		●					●		●			
16-033	Catherine Riddle	Investigation of Gadolinium Nanocrystal Gels for Scintillator Use in Neutron Detection	●						●	●				●
16-036	Muhammad Abir	Neutron Microscope to Enable High-Resolution Neutron Tomography at INL							●					●
16-040	Vivek Agarwal	Integration of Prognostic Techniques and Probabilistic Safety Assessment for Online Risk Monitoring			●			●	●				●	

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Project Number	PI Name	Title	Department of Homeland Security	Department of Defense	DOE Office of Energy Efficiency and Renewable Energy	DOE Office of Environmental Management	DOE Office of Environmental, Safety, and Health	DOE Office of Fossil Energy	DOE-NE	National Nuclear Security Administration	DOE Office of Naval Reactors	DOE Office of Civilian Radioactive Waste Management	DOE-Office of Electricity Delivery and Energy Reliability	DOE Office of Science
16-046	Assel Aitkaliyeva	Development of a Synergistic Approach to Study Irradiated Materials Using Coupled Experiments and Simulation							●					
16-050	Sebastien Teyssyre	Stress Corrosion Cracking Testing in Supercritical Carbon Dioxide							●					●
16-055	Cody Permann	Capability Extension for Multi-Scale, Multi-Application Development within the Multi-Physics Object-Oriented Simulation Environment		●	●	●		●	●		●			●
16-058	Cody Permann	Predicting Radiation-Induced Microstructural Change via Implementation and Validation of Multi-Scale Cluster Dynamics in MOOSE		●					●	●	●		●	●
16-070	Aaron Craft	Characterization of Neutron Beamlines at NRAD							●					
16-071	Aaron Craft	Evaluation of Advanced Digital Neutron Imaging Systems for PIE of Nuclear Fuel							●					
16-081	Ronald Heaps	Modeling Thermite Reactions	●						●					
16-085	Christopher Zarzana	Production of Fluoroanion Targets for Accelerator Mass Spectrometry							●					
16-096	Katya Le Blanc	Supporting Operator Performance and Situation Awareness in Highly Automated Nuclear Power Plants			●				●				●	



Project Number	PI Name	Title	Department of Homeland Security	Department of Defense	DOE Office of Energy Efficiency and Renewable Energy	DOE Office of Environmental Management	DOE Office of Environmental, Safety, and Health	DOE Office of Fossil Energy	DOE-NE	National Nuclear Security Administration	DOE Office of Naval Reactor	DOE Office of Civilian Radioactive Waste Management	DOE-Office of Electricity Delivery and Energy Reliability	DOE Office of Science
16-098	Mathew Snow	Nuclear Nonproliferation Applications of $^{14}\text{C}$ Analyses by Accelerator Mass Spectrometry	•							•				
16-106	Mary Klett	Risk Analysis Method Integrating Physical, Cyber and Infrastructure Dependencies	•										•	
16-129	Shelly Li	ICS-CAPE: Industrial Control Systems – Cyber Attacks and their Physical Effects							•	•				
16-133	Briam Johnson	Secure SCADA Communications System	•		•				•				•	
16-149	Joshua Daw	In-Core Qualification of Developmental Instrumentation		•					•		•			
16-152	Kurt Derr	Wireless Radiofrequency Signal Identification and Protocol Reverse Engineering	•										•	•
16-176	Ting He	Development of Direct Carbon Fuel Cells			•			•						•
16-187	Chao Jiang	Micro-Scale Technique to Evaluate Grain Boundary Cohesion of Irradiated Alloys						•	•	•				•
16-215	Ting He	Electrochemical Manufacturing Processes			•			•	•					•
16P6-002FP	Chinmoy Baroi	Kinetic-Based Scale-Up Science for an Energy Efficient Route to Ethylene			•		•	•						•
16P6-003FP	James Sterbentz	Phenomena Identification and Ranking Table Technique Applied to the MegaPower Heat Pipe Reactor Concept		•					•					

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16P6-007FP	Robert T Smith	Consequence-Driven Cyber-Informed Engineering Methodology Pilot	•						•	•			•	
<b>FY 2016 Projects<sup>+</sup></b>			<b>17</b>	<b>9</b>	<b>29</b>	<b>12</b>	<b>4</b>	<b>14</b>	<b>54</b>	<b>18</b>	<b>8</b>	<b>3</b>	<b>25</b>	<b>31</b>
<b>FY 2016 Investments (\$M)<sup>*</sup></b>			<b>2.7</b>	<b>2.2</b>	<b>8.0</b>	<b>3.0</b>	<b>0.8</b>	<b>4.0</b>	<b>13.0</b>	<b>3.2</b>	<b>1.9</b>	<b>0.8</b>	<b>5.4</b>	<b>8.3</b>

<sup>+</sup> Project total exceeds 71 projects funded in 2016 because multiple beneficiaries per project are allowed.

<sup>\*</sup> Project total funding exceeds the approximately \$17.8M FY 2016 LDRD budget because multiple beneficiaries are allowed for each project.

## **Appendix B**

### **Publications List for 2015 and 2016**



## Appendix B

### Publications List for 2015 and 2016

#### FY 2015

	Journal Citation	Impact Factor	LDRD No.	Initiative	Collaborator Affiliation State/Country
1	Bair, Jacob, Mohsen Asle Zaeem, and Michael R. Tonks, 2015, "A Review on Hydride Precipitation in Zirconium Alloys," <i>Journal of Nuclear Materials</i> Vol. 466, pp. 12-20.	2.199	13-032	ANE	Missouri University Science and Technology, Missouri
2	Chen, Jun and Humberto E. Garcia, 2016, "Economic Optimization of Operations for Hybrid Energy Systems under Variable Markets," <i>Applied Energy</i> Vol. 177, pp. 11-24.	5.746	13-065	CED	No external collaborators
3	Chen, X., L. M. Xiong, A. Chernatynskiy, and Y. P. Chen, 2014, "A Molecular Dynamics Study of Tilt Grain Boundary Resistance to Slip and Heat Transfer in Nanocrystalline Silicon," <i>Journal of Applied Physics</i> Vol. 116, pp. 244309.	2.101	13-105	ANE	University of Florida, Florida Iowa State University, Iowa
4	Chernatynskiy, Aleksandr, Xian-Ming Bai, and Jian Gan, 2016, "Systematic Investigation of the Misorientation-and Temperature-Dependent Kapitza Resistance in $\text{CeO}_2$ ," <i>International Journal of the Heat and Mass Transfer</i> Vol. 99, pp. 461-469.	2.857	13-105	ANE	University of Florida, Florida
5	Clark, Billy, Jena McCollum, Michelle L. Pantoya, Ronald J. Heaps, Michael A. Daniels, Emily M. Hunt, Trent J. Kelly, and Brenton F. Allen, 2015, "Development of Flexible, Free-standing, Thin Films for Additive Manufacturing and Localized Energy Generation," <i>AIP Advances</i> Vol. 5, Issue 8, pp. 087128.	1.444	14-035	CED	Texas Tech University, Texas

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	<b>Journal Citation</b>	<b>Impact Factor</b>	<b>LDRD No.</b>	<b>Initiative</b>	<b>Collaborator Affiliation State/Country</b>
6	Collins, E. S., B. R. Skelton, M. L. Pantoya, F. Irin, M. J. Green, and M. A. Daniels, 2015, “Ignition Sensitivity and Electrical Conductivity of an Aluminum Fluoropolymer Reactive Material with Carbon Nanofillers,” <i>Combustion and Flame</i> , Vol. 162, Issue 4, pp. 1417–1421.	4.168	12-066	ANE	Texas Tech University, Texas Texas A&M, Texas
7	Dufek, Eric J., Michael Picker, and Lucia M. Petkovic, 2016, “Processing Impact on Composite Si/Graphite Electrodes,” <i>Journal of Applied Electrochemistry</i> Vol. 46, Issue 3, pp. 359-367.	2.223	13-027	CED	No external collaborators
8	Heaps, Ronald J., 2015, “Synthesis and Characterization of Flexible, Free-standing, Energetic Thin Films,” <i>42nd International Conference on Metallurgical Coatings and Thin Films</i> Vol. 284, pp. 422-426.	1.761	14-035	CIP	Texas Tech University, Texas West Texas A&M, Texas
9	Huang, H. B. Spencer, and J. Hales, 2014, “Discrete Element Method for Simulation of Early-Life Thermal Fracturing Behavior in Ceramic Nuclear Fuel Pellets,” <i>Nuclear Engineering and Design</i> Vol. 278, pp. 515–528.	0.967	13-071	ANE	No external collaborators
10	Kadioglu, Samet Y, 2016, “An Essentially Non-Oscillatory Spectral Deferred Correction Method for Hyperbolic Problems,” <i>Journal of Computational Methods</i> Vol. 13, Issue 3, pp. 165007.	1.345	14-104	ANE	Yildiz Technical University, Istanbul, Turkey
11	Kaur, M., Y. Qiang, W. L. Jiang, C. Pearce, and J. S. McCloy, 2014, “Magnetization Measurements and XMCD Studies on Ion Irradiated Iron Oxide and Core-Shell Iron/Iron-Oxide Nanomaterials,” <i>IEEE Transactions on Magnetics</i> Vol. 50, Issue 11, pp. 4800305.	1.277	NA	ANE	University of Idaho, Idaho Pacific Northwest National Laboratory, Washington University of Manchester, Lancs, England (United Kingdom) Washington State University, Washington



	Journal Citation	Impact Factor	LDRD No.	Initiative	Collaborator Affiliation State/Country
12	Klaehn, John R., Christopher J. Orme, and Eric S. Peterson, 2016, "Blended Polybenzimidazole and Melamine-co-Formaldehyde Thermosets," <i>Chemical Engineer Journal</i> Vol. 515, pp. 1–6.	5.31	08-043	CED	No external collaborators
13	Laraway, Stephen Andrew, Behrouz Farhang-Boroujeny, and Hussein Moradi, 2015, "HF Band Filter Bank Multi-Carrier Spread Spectrum," <i>IEEE Communications Society MILCOM 2015</i> , DOI: 10.1109/MILCOM.2015, pp. 7357648.	6.524	13-093	CIP	University of Utah, Utah
14	Lu, C., C. Zhang, H. Hunag, and T. C. Johnson, 2015, "Monitoring CO <sub>2</sub> Sequestration Into Deep Saline Aquifer and Associated Salt Intrusion Using Coupled Multiphase Flow Modeling and Time-lapse Electrical Resistivity Tomography," <i>Greenhouse Gases-Science and Technology</i> Vol. 5, Issue 1, pp. 34-49.	1.92	11-045	CED	Chinese Academy of Geology Sciences, Institute of Hydrology and Environmental Geology, China Rutgers State University, New Jersey Pacific Northwest National Laboratory, Washington
15	Ma, W., F. W. Herbert, S. D. Senanayake, and B. Yildiz, 2015, "Non-Equilibrium Oxidation States of Zirconium During Early Stages of Metal Oxidation," <i>Applied Physics Letters</i> Vol. 106, pp. 101603.	3.142	12-026	ANE	Massachusetts Institute of Technology, Massachusetts Brookhaven National Laboratory, New York
16	Majid, Arslan, Behrouz Farhang, and Hussein Moradi, "Secure Information Transmission in Filter Bank Multi-Carrier Spread Spectrum Systems," <i>IEEE Communications Society MILCOM 2015</i> , DOI: 10.1109/MILCOM.2015, pp. 7357457.	6.524	13-093	CIP	University of Utah; Utah

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	<b>Journal Citation</b>	<b>Impact Factor</b>	<b>LDRD No.</b>	<b>Initiative</b>	<b>Collaborator Affiliation State/Country</b>
17	Masuda, K., E. I. Vaughan, L. Arissian, J. P. Hendrie, J. Cole, J. -C. Diels, and A. A. Hecht, 2015, “Novel Techniques for High Precision Refractive Index Measurements, and Application to Assessing Neutron Damage and Dose in Crystals,” <i>Nuclear Instruments and Methods in Physics Research</i> Vol. 784, pp. 198–201.	1.2	14-012	ANE	University of New Mexico, New Mexico
18	McNally, Joshua S., Bruce Noll, Christopher J. Orme, and Aaron D. Wilson, 2015, “Density Functional Theory Analysis of the Impact of Steric Interaction on the Function of Switchable Polarity Solvents,” <i>Journal of Chemical Physics B</i> , Vol. 119, Issue 22, pp. 6766–6775.	3.187	14-079	CED	Bruker AXS, Inc. Wisconsin
19	Mincher, Bruce J., Chien M. Wai, Robert V. Fox, Donna L. Baek, Clive Yen, and Mary E. Case, 2016, “The Separation of Lanthanides and Actinides in Supercritical Fluid Carbon Dioxide,” <i>Journal of Radioanalytical and Nuclear Chemistry</i> Vol. 307, pp. 2543–2547.	0.983	11-006	ANE	University of Idaho, Idaho
20	Mincher, Bruce J., Martin Precek, and Alena Paulenova, 2016, “The Redox Chemistry of Neptunium in Gamma-irradiated Aqueous Nitric Acid in the Presence of an Organic Phase,” <i>Journal of Radioanalytical and Nuclear Chemistry</i> Vol. 308, pp. 1005.	0.983	12-010	ANE	Oregon State University, Oregon Academy of Sciences of the Czech Republic; Prague, Czech Republic.
21	Moradi, Hussein and Behrouz Farhang-Boroujeny, 2016, “OFDM Inspired Waveforms for 5G,” <i>IEEE Communications Surveys and Tutorials</i> Vol. 18, No. 4, pp. 2474–2492.	9.22	13-093	CIP	University of Utah, Utah
22	Orme, Christopher J. and Aaron D. Wilson, 2015, “1-Cyclohexylpiperidine as a Thermolytic Draw Solute for Osmotically Driven Membrane Processes,” <i>Desalination</i> Vol. 371, pp. 126–133.	4.412	14-079	CED	No external collaborators

	<b>Journal Citation</b>	<b>Impact Factor</b>	<b>LDRD No.</b>	<b>Initiative</b>	<b>Collaborator Affiliation State/Country</b>
23	Poper, Kade, Billy Clark, Michelle L. Pantoya, Ron Heaps, and Michael Daniels, 2015, "Desensitizing Ignition of Energetic Materials When Exposed to Accidental Fire," <i>Fire Safety Journal</i> , Vol. 76, pp. 39-43.	0.936	14-035	CIP	Texas Tech University, Texas
24	Poper, K. H., E. S. Collins, M. L. Pantoya, and M. A. Daniels, 2014, "Controlling the Electrostatic Discharge Ignition Sensitivity of Composite Energetic Materials Using Carbon Nanotube Additives," <i>Journal of Electrostatics</i> Vol. 72, Issue 5, pp. 428-432.	1.286	14-035	CIP	Texas Tech University, Texas
25	Reimund, Kevin K., Benjamin J. Coscia, Jason T. Arena, Aaron D. Wilson, and Jeffrey R. McCutcheon, 2016, "Characterization and Membrane Stability Study for the Switchable Polarity Solvent N, N-Dimethylcyclohexylamine as a Draw Solute in Forward Osmosis," <i>Journal of Membrane Science</i> Vol. 501, pp. 93-99.	5.557	14-079	CED	University of Connecticut, Connecticut
26	Riddle, Catherine, Kenneth Czerwinski, Eunja Kim, Patricia Paviet, Philippe Weck, Frederic Poineau, and Steven Conradson, 2016, "Characterization of Pentavalent and Hexavalent Americium Complexes in Nitric Acid Using X-ray Absorption Fine Structure Spectroscopy and First-Principles Modeling," <i>Journal of Radioanalytical and Nuclear Chemistry</i> Vol. 309, Issue 3, pp. 1087-1095.	0.983	11-005	ANE	University of Nevada, Nevada DOE, Washington D.C. Sandia National Laboratory, New Mexico Synchrotron Soleil Laboratory, Saint Aubin, France

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	<b>Journal Citation</b>	<b>Impact Factor</b>	<b>LDRD No.</b>	<b>Initiative</b>	<b>Collaborator Affiliation State/Country</b>
27	Steelman, Ryan, Billy Clark, Michelle L. Pantoya, and Michael Daniels, 2015, “Desensitizing Nano Powders to Electrostatic Discharge Ignition,” <i>Journal of Electrostatics</i> Vol. 76, pp. 102-107.	1.286	14-035	CIP	Texas Tech University, Texas
28	Teague, M. C., B. S. Fromm, M. R. Tonks, and D. P. Field, 2014, “Using Coupled Mesoscale Experiments and Simulations to Investigate High Burn-Up Oxide Fuel Thermal Conductivity,” <i>Journal of the Minerals, Metals, and Materials Society</i> Vol. 66, pp. 2569-2577, DOI:10.1007/s11837-014-1160-3.	1.798	12-031	ANE	Washington State University, Washington
29	Tripathy, P. K., L. A. Wurth, E. J. Dufek, T. Y. Gutknecht, N. J. Gese, P. A. Hahn, S. M. Frank, G. L. Fredrickson, and J. S. Herring, 2014, “Aluminum Electroplating On Steel From A Fused Bromide Electrolyte,” <i>Surface and Coatings Technology</i> Vol. 258, pp. 652–663.	2.139	13-014	CED	ZAF Energy Systems Inc., Montana Chalmers, Gothenburg, Sweden
30	Wilson, Aaron D., Kevin K. Reimund, and Jeffrey R. McCutcheon, 2015, “Thermodynamic Analysis of Energy Density in Pressure Retarded Osmosis: The Impact of Solution Volumes and Costs,” <i>Journal of Membrane Science</i> Vol. 487, pp. 240-248.	5.557	14-079	CED	University of Connecticut, Connecticut
31	Wilson, A. D. and C. J. Orme, 2015, “Concentration Dependent Speciation and Mass Transport Properties of Switchable Polarity Solvents,” <i>RSC Advances</i> Vol. 5, pp. 7740-7751.	3.289	14-079	CED	No external collaborators
32	Zalupski, P. R., R. McDowell, and G. Dutech, 2014, “The Adsorption of Gold, Palladium, and Platinum from Acidic Chloride Solutions on Mesoporous Carbons,” <i>Solvent Extraction and Ion Exchange</i> Vol. 32, Issue 7, pp. 737-748.	1.404	12-056	ANE	No external collaborators

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CIP = critical infrastructure

## FY 2016

	Journal Citation	Impact Factor	LDRD No.	Initiative	Collaborator Affiliation State/Country
1	Agarwal, Vivek and James A. Smith, "Near Real-Time In-Pile Acoustic Measurement Infrastructure at the Advanced Test Reactor," <i>ANS Nuclear Technology</i> , in review.	0.59	15-040	ANE	No external collaborators
2	Aston, J. E., W. A. Apel, B. D. Lee, D. N. Thompson, J. A. Lacey, D. T. Newby, D. W. Reed, and V. S. Thompson, 2016, "Degradation of Phenolic Compounds by the Lignocellulose Deconstructing Thermoacidophilic Bacterium Alicyclobacillus Acidocaldarius," <i>Journal of Industrial Microbiology and Biotechnology</i> Vol. 43, Issue 1, pp. 12-23.	2.745	12-051	CED	Pacific Northwest National Laboratory, Washington
3	Bao, Han, Haihua Zhao, Hongbin Zhang, Ling Zou, Phil Sharpe, and Nam Dinh, "Safe Reactor Depressurization Windows for BWR Mark I Station Blackout Accident Management Strategy," <i>Annals of Nuclear Energy</i> , in review.	1.174	16-026	ANE	No external collaborators
4	Biner, S. B., W. F. Rao, and Y. F. Zhang, 2016, "The Stability of Precipitates and the Role of Lattice Defects in Fe-1at% Cu-1at%Ni-1at%Mn Alloy: A Phase-Field Model Study," <i>Journal of Nuclear Materials</i> Vol. 468, pp. 9-16.	2.199	13-013	ANE	No external collaborators
5	Carroll, M. C, M. R. Tonks, T. M. Lillo, B. S. Fromm, D. C. Haggard, T. C. Morris, W. D. Swank, T. L. Trowbridge, and L. J. Carroll, "Characteristics of Interconnected Delta-Hydride Precipitates in Zr," <i>Metallurgical and Materials Transactions E</i> , accepted with revisions.	1.749	13-032	ANE	No external collaborators

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	<b>Journal Citation</b>	<b>Impact Factor</b>	<b>LDRD No.</b>	<b>Initiative</b>	<b>Collaborator Affiliation State/Country</b>
6	Clark, B. R., M. L. Pantoya, E. M. Hunt, T. J. Kelly, B. F. Allen, R. J. Heaps, and M. A. Daniels, 2015, “Synthesis and Characterization of Flexible, Free-standing, Energetic Thin Films,” <i>Surface and Coatings Technology</i> Vol. 284, pp. 422–426.	2.139	14-035	CIP	Texas Tech University, Texas West Texas A&M University, Texas
7	Dufek, E. J., M. Picker, and L. M. Petkovic, 2016, “Density Impact On Performance of Composite Si/Graphite Electrodes,” <i>Journal of Applied Electrochemistry</i> Vol. 46, Issue 3, pp. 359–367.	2.223	13-027	CED	Montana State University, Montana
8	Dufek, Eric J., John R. Klaehn, Joshua S. McNally, Harry W. Rollins, and David K. Jamison, 2016, “Use of Phosphoranimines to Reduce Carbonates In Li-ion Battery Electrolytes,” <i>Electrochimica Acta</i> Vol. 209, pp. 36-43.	4.803	15-125	CED	No external collaborators
9	Ellis, R. J. and J. Rapp, 2015, “Neutron-Irradiated Samples as Test Materials for MPEX,” <i>Fusion Science and Technology</i> Vol. 68, No. 4, pp. 750-757.	0.799	NA	ANE	Oak Ridge National Laboratory, Tennessee
10	Gering, Kevin L., “Prediction of Electrolyte Conductivity: Results from a Generalized Molecular Model Based on Ion Solvation and a Chemical Physics Framework,” <i>Electrochimica Acta</i> , in review.	4.803	15-140	CED	No external collaborators
11	Gering, Kevin L. and Joshua S. McNally, “Ion Solvation Metrics and Predictive Methods based on Contributions from Density Functional Theory and an Advanced Chemical Physics Approach,” <i>Electrochimica Acta</i> , In Review.	4.803	15-140	CED	No external collaborators
12	Glazoff, Michael and Eric Dufek, 2016, “Morphological Analysis and Synthesis for Understanding Electrode Microstructure Evolution as a Function of Applied Charge/Discharge Cycles,” <i>Applied Physics A</i> Vol. 122, pp. 894-906.	1.444	13-027	CED	No external collaborators



	Journal Citation	Impact Factor	LDRD No.	Initiative	Collaborator Affiliation State/Country
13	Hoggan, Rita, Larry Zuck, W. Roger Cannon, and Paul A. Lessing, 2016, "Processing of Surrogate Nuclear Fuel Pellets for Better Dimensional Control with Dry Bag Isostatic Pressing," <i>Journal of Nuclear Materials</i> Vol. 482, pp. 34-41.	2.199	11-051	ANE	Rutgers University, New Jersey
14	Klaehn, John R., Christopher J. Orme, and Eric S. Peterson, 2016, "Blended Polybenzimidazole and Melamine-Co-Formaldehyde Thermosets," <i>Journal of Membrane Science</i> Vol. 515, pp. 1-6.	5.557	08-043	ANE	No external collaborators
15	Lee, Brady, "Concurrent Metabolism of Pentose and Hexose Sugars by the Polyextremophile Alicyclobacillus Acidocaldarius," <i>Journal of Industrial Microbiology and Biotechnology</i> , in review.	2.745	07-123	CED	No external collaborators
16	Liu, Ren, Anurag Srivastava, Mayank Panwar, Manish Mohanpurkar, and Rob Hovsapien, 2017, "Geographically Distributed Real-Time Digital Simulations Using Linear Prediction," <i>International Journal of Electrical Power and Energy Systems</i> , Vol. 84, pp. 308-317.	2.587	15-135	CED	Washington State University, Washington Colorado State University, Colorado
17	Meher, S., L. J. Carroll, M. C. Carroll, and T. M. Pollock, 2016, "Solute Partitioning in Multi-component $\gamma/\gamma'$ Co-Ni-base Superalloys with Near-Zero Lattice Misfit," <i>Scripta Materialia</i> Vol. 113, pp. 185-189.	3.305	14-078	ANE	University of California, Santa Barbara, California
18	Mincher, B. J., M. Precek, and A. Paulenova, 2016, "The Redox Chemistry of Neptunium in Gamma-irradiated Aqueous Nitric Acid in the Presence of an Organic Phase," <i>Journal of Radioanalytical and Nuclear Chemistry</i> , Vol. 308, pp. 1005-1009.	0.983	12-010	ANE	Oregon State University, Oregon Academy of Sciences in the Czech Republic, Prague, Czech Republic

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	<b>Journal Citation</b>	<b>Impact Factor</b>	<b>LDRD No.</b>	<b>Initiative</b>	<b>Collaborator Affiliation State/Country</b>
19	Misenheimer, Corey T. and Dr. Stephen D. Terry, 2016, “Modeling Hybrid Nuclear Systems with Chilled-Water Storage,” <i>ASME Journal of Energy Resources Technology</i> Vol. 139, Issue 1, pp. 012002.	1.347	15-039	ANE	North Carolina State University, North Carolina
20	Mitchell, Jessica J., Nancy F. Glenn, Matthew O. Anderson, and Ryan Hruska, 2016, “Flight Considerations and Hyperspectral Image Classifications for Dryland Vegetation Management from a Fixed-wing UAS,” <i>Environmental Management and Sustainable Development</i> , Macrothink Institute, ISSN 2164-7682, Vol. 5, No. 2.	0.739	09-019	CED	Appalachian State University, North Carolina Boise State University, Idaho
21	Moody, Alex, Jerry Fairley, and Mitchell Plummer, 2016, “Stochastic Exploration and the Geologic Context of Enhanced Geothermal System Viability on the Snake River Plain, Idaho,” <i>Idaho Geological Society of America Special Papers: Geothermal Energy: An Emerging Resource</i> , GSA Special Papers, Vol. 519, pp. 115-136.	4.398	13-068	CED	University of Idaho, Idaho
22	Morgan, Kevin, Noleen Maguire, Rebecca Fushimi, John T. Gleaves, Alexandre Goguet, Michael P. Harold, Evgenii V. Kondratenko, Unmesh Menon, Yves Schuurman, and Gregory S. Yablonsky, “Temporal Analysis of Products (TAP): A Hidden Jewel in the Crown of Heterogeneous Catalysis Kinetic Analysis, <i>ACS Catalysis</i> , in review.	9.307	15-146	CED	No external collaborators

	Journal Citation	Impact Factor	LDRD No.	Initiative	Collaborator Affiliation State/Country
23	Nielsen, J., A. Tokuhito, R. Hiromoto, and L. Tu, 2015, "Branch-and-Bound Algorithm Applied to Uncertainty Quantification of a Boiling Water Reactor Station Blackout," <i>Nuclear Engineering and Design</i> Vol. 295, pp. 283-304.	0.967	13-040	ANE	University of Idaho, Idaho
24	Osorio, Julian D., Rob Hovsopian, and Juan C. Ordonez, 2016, "Dynamic Analysis of Concentrated Solar Supercritical CO <sub>2</sub> -based Power Generation Closed-Loop Cycle," <i>Applied Thermal Engineering</i> Vol. 93, pp. 920-934.	3.043	15-135	CED	Florida State University, Florida
25	Panwar, Mayank, Rob Hovsopian, and Siddharth Suryanarayanan, "A Performance Metric for Reserve Management in Day-Ahead Dispatch of Electric Microgrids," <i>Applied Energy</i> , in review.	5.746	15-135	CED	No external collaborators
26	Panwar, Mayank, Siddharth Suryanarayanan, and Rob Hovsopian, "A Multi-Criteria Decision Analysis-Based Approach for Dispatch of Electric Microgrids," <i>IEEE - Transactions</i> , in review.	1.277	15-135	CED	No external collaborators
27	Pasebani, S., I. Charit, D. P. Butt, J. I. Cole, Y. Q. Wu, and J. Burns, 2016, "Sintering Behavior of Lanthana-Bearing Nanostructured Ferritic Steel Consolidated via Spark Plasma Sintering," <i>Advanced Engineering Materials</i> Vol. 18, Issue 2, pp. 324-332.	1.82	10-056	ANE	University of Idaho, Idaho Boise State University, Idaho Center for Advanced Energy Studies, Idaho
28	Pasebani, S., I. Charit, Y. Q. Wu, J. Burns, K. N. Allahar, D. P. Butt, J. I. Cole, and S. F. Alsagabi, 2016, "Lanthana-bearing Nanostructured Ferritic Steels Via Spark Plasma Sintering," <i>Journal of Nuclear Materials</i> Vol. 470, pp. 297-306.	2.199	10-056	ANE	University of Idaho, Idaho Boise State University, Idaho Center for Advanced Energy Studies, Idaho
29	Pawar, Gorakh, Hai Huang, and Adri Duin, "Evolution of Subnanometer Scale Features in Realistic Kerogen Matrix," <i>Journal of Applied Physics</i> , in review.	2.101	15-128	ANE	No external collaborators

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	<b>Journal Citation</b>	<b>Impact Factor</b>	<b>LDRD No.</b>	<b>Initiative</b>	<b>Collaborator Affiliation State/Country</b>
30	Pawar, Gorakh, Hai Huang, and Ilija Miskovic, "The Multiphase Simulation in Realistic Nanometer and Subnanometer Pores Found in Kerogen Matrix," <i>Energy and Fuels</i> , in review.	2.835	15-128	ANE	No external collaborators
31	Pawar, Gorakh, Hai Huang, Paul Meakin, and Ilija Miskovic, "Molecular Dynamics Simulation and Subnanometer Scale Characterization of Kerogen with a General-Purpose DREIDING Force Field," <i>Energy and Fuels</i> , in review.	2.835	15-128	ANE	No external collaborators
32	Reimund, K. K., B. J. Coscia, J. T. Arena, A. D. Wilson, and J. R. McCutcheon, 2016, "Characterization and Membrane Stability Study for the Switchable Polarity Solvent N,N-Dimethylcyclohexylamine as a Draw Solute in Forward Osmosis," <i>Journal of Membrane Science</i> Vol. 501, pp. 93–99.	5.557	14-079	CED	University of Connecticut, Connecticut
33	Smith, S. A., E. Hughes, E. R. Coats, C. K. Brinkman, A. G. McDonald, J. R. Harper, K. Feris, and D. Newby, 2016, "Toward Sustainable Dairy Waste Utilization: Enhanced VFA and Biogas Synthesis via Upcycling Algal Biomass Cultured on Waste Effluent," <i>Journal of Chemical Technology and Biotechnology</i> Vol. 91, Issue 1, pp. 113-121.	2.738	NA	CED	University of Idaho, Idaho Boise State University, Idaho
34	Tan, Shuai, Shibely Saha, Rebecca R. Fushimi, Lucun Wang, and Dongmei Li, "Experimentally Probing Ligand-Strain Effect via a Novel Catalyst Platform - Full Manuscript," <i>Journal of the American Chemical Society</i> , in review.	13.038	15-146	CED	No external collaborators
35	Tang, Mingchen, Ting He, and Maohong Fan, "Coal-Derived Carbon Materials: A Modern Approach to Unlock Nature's Complexity and Treasure," <i>Progress in Materials Science</i> , in review.	31.083	16-176	CED	No external collaborators

	<b>Journal Citation</b>	<b>Impact Factor</b>	<b>LDRD No.</b>	<b>Initiative</b>	<b>Collaborator Affiliation State/Country</b>
36	Wang, Lu-Cun, Michelle Personic, Rebecca Fushimi, Cynthia M. Friend, and Robert J. Madix, "Activation of Dioxygen and Methanol at Active Sites on Silver/Gold Nanoporous Catalysts," <i>The Journal of Physical Chemistry Letters</i> , in press.	4.509	15-146	CED	No external collaborators
37	Williams, A. N., M. Pack, and S. Phongikaroon, 2015, "Separation of Strontium and Cesium from Ternary and Quarternary Lithium Chloride-Potassium Chloride Salts Via Melt Crystallization," <i>Nuclear Engineering and Technology</i> Vol. 47, Issue 7, pp. 867-874.	0.792	NA	ANE	Virginia Commonwealth University, Virginia University of Idaho, Idaho
38	Xia, Y., G. Jan, H. Huang, I. Miskovic, P. Meakin, and M. Deo, 2016, "Many-Body Dissipative Particle Dynamics Modeling of Fluid Flow in Fine-Grained Nanoporous Shales," <i>Physics of Fluids</i> , in review.	2.017	15-128	CED	No external collaborators
39	Xia, Yidong, David Andrs, Richard C. Martineau, and A Hermite, 2016, "WENO-Based Slope Limiter for Finite Volume Method on Unstructured Grids," <i>Journal of Computational Physics</i> , in review.	2.556	14-104	ANE	No external collaborators
40	Xia, Yidong, Jan Goral, Hai Huang, Ilija Miskovic, and Paul Meakin, 2016, "Coupled Digital Rock Physics and Dissipative Particle Dynamics Modeling and Simulation of Multiphase Fluid Flow in Organic-Rich Heterogeneous Shale Reservoir Pore System," <i>Physics of Fluids</i> , in review.	2.017	15-128	CED	No external collaborators
41	Zhang, Huijin, G. Rocklan McDowell, Leigh R. Martin, and You Qiang, 2016, "Selective Extraction of Heavy and Light Lanthanides from Aqueous Solution by Advanced Magnetic Nanosorbents," <i>ACS Applied Materials and Interfaces</i> Vol. 8, Issue 14, pp. 9532–9531.	2.017	13-033	ANE	University of Idaho, Idaho

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	<b>Journal Citation</b>	<b>Impact Factor</b>	<b>LDRD No.</b>	<b>Initiative</b>	<b>Collaborator Affiliation State/Country</b>
42	Zhang, Y. F., X. M. Bai, J. G. Yu, M. R. Tonks, M. J. Noordhoek, and S. R. Phillpot, 2016, “Homogeneous Hydride Formation Path in Alpha-Zr: Molecular Dynamics Simulations with the Charge-optimized Many-body Potential,” <i>Acta Materialia</i> Vol. 111, pp. 357-365.	5.058	14-026	ANE	Penn State University, Pennsylvania University of South Carolina, South Carolina University of Florida, Florida
43	Zhou, Jing, Hai Huang, John McLennan, and Milind Deo, “A Dual-Lattice Discrete Element Model to Understand Hydraulic Fracturing in a Naturally Fractured System,” <i>Hydraulic Fracturing Journal</i> , in review.	2.045	15-128	CED	No external collaborators
44	Zhou, Jing, Hai Huang, Shu Jiang, and Milind Deo, “A New Physics-Based Modeling of Multiple Non-Planar Hydraulic Fracture Propagation,” <i>Society of Petroleum Engineers Journal</i> , in review.	1.137	15-128	CED	No external collaborators

ANE = advancing nuclear energy

CED = clean energy development

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## **Appendix C**

### **List of Collaborators**



## Appendix C

### List of Collaborators

#### University Collaborations

University	Projects	Scope
Boise State University	<p>14-086: Development of a Microgrid/Smartgrid Testbed for ESL and Super Lab Initiative, with Load Variability Characterization and Control for Renewable Energy Integration</p> <p>15-128: Microstructural Evolution and Mesoscale Coupled Flow-Reaction-Fracturing Processes in Organic Rich Nanoporous Shales</p> <p>15-140: Expanding the Utility of Advanced Chemical Physics Models for Electrolytes</p> <p>16-176: Development of Direct Carbon Fuel Cells</p> <p>16-187: Micro-Scale Technique to Evaluate Grain Boundary Cohesion of Irradiated Alloys</p>	<p>Determine the contribution of variability from renewable generation and various load types and how management, regulation, and control of grid systems can benefit through optimized integration, improved use-case control algorithms, and testing of new and existing load, storage, and generation resource technologies. Modeling and simulation to develop better understanding of the fundamental physics that controls the behaviors of organic-rich nanoporous shales and the fluids within them. Develop fast computational tools that can economically accelerate selection and characterization of advanced electrolyte systems; direct carbon fuel cell; and research and development in nuclear energy structural and cladding materials.</p>
Carnegie Mellon	16-106: ICS-CAPE: Industrial Control Systems – Cyber Attacks and their Physical Effects	Identify and understand the physical impacts of a cyber-attack on an industrial control system.
Colorado State University	15-135: Dynamic Simulations for Large Scale Electric Power Networks in Real-Time Environment using Multiple RTDS	Test and evaluate necessary components for geographically distributed real-time simulation using real-time simulators and interconnectivity between different locations

University	Projects	Scope
Florida State University	14-086: Development of a Microgrid/Smartgrid Testbed for ESL and Super Lab Initiative, with Load Variability Characterization and Control for Renewable Energy Integration 15-135: Dynamic Simulations for Large Scale Electric Power Networks in Real-Time Environment using Multiple RTDS	Determine the contribution of variability from renewable generation and various load types and how management, regulation and control of grid systems can benefit through optimized integration, improved use-case control algorithms, and testing of new and existing load, storage, and generation resource technologies. Test and evaluate necessary components for geographically distributed real-time simulation using real-time simulators and interconnectivity between different locations.
Idaho State University	14-086: Development of a Microgrid/Smartgrid Testbed for ESL and Super Lab Initiative, with Load Variability Characterization and Control for Renewable Energy Integration 15-145: Advanced Neutron and X-Ray Imaging at TREAT 16-070: Characterization of Neutron Beamlines at NRAD	Determine the contribution of variability from renewable generation and various load types and how management, regulation, and control of grid systems can benefit through optimized integration, improved use-case control algorithms, and testing of new and existing load, storage, and generation resource technologies. Develop advanced imaging options for the future TREAT experimental program. Characterize the two neutron beams at Hot Fuel Examination Facility and the East and North Radiography Stations to provide users with detailed information about these beams, including the beam flux, spatial distribution, energy spectrum, divergence/collimation, and gamma content.
Illinois Institute of Technology	14-098: Irradiation Effects in Uranium Dioxide	Investigating defect production, evolution, and fission product transport under irradiation and revealing their physical mechanisms.

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University	Projects	Scope
Kyoto University	16-085: Production of Fluoroanion Targets for Accelerator Mass Spectrometry	Detection of nuclides of the actinide elements using accelerator mass spectrometry for an order of magnitude increase in sensitivity for actinide analysis.
Massachusetts Institute of Technology	15-039: Transient Modeling of Integrated Nuclear Energy Systems with Thermal Energy Storage and Component Aging and Preliminary Model Validation via Experiment 15-060: Development of Efficient TREAT Modeling Capabilities with Graphite Data Improvement 16-036: Neutron microscope to enable high-resolution neutron tomography at INL 16-058: Predicting Radiation-Induced Microstructural Change via Implementation and Validation of Multiscale Cluster Dynamics in MOOSE 16-149: In-core Qualification of Developmental Instrumentation	Focus on transient modeling of integrated, hybrid energy systems that would incorporate nuclear and renewable generators, electricity production, an industrial process, and thermal energy storage. Research efforts to support modeling and experimentation related TREAT. Design focusing mirrors, sometimes called Wolter mirrors, for the neutron radiography application at INL's NRAD. Study the radiation defect cluster distribution and how it contributes to the successful design of various radiation-resistant materials. This is not limited to nuclear applications and predicting their stability under irradiation. Perform ex-core characterization of several sensors under development for in-core use.
Montana Tech	16-003: Recycling of Tantalum-Containing Waste Materials to Recover Tantalum Metal	Develop a high-temperature electrochemical process with a view to reclaiming tantalum metal from its primary and secondary resources.
North Carolina State University	15-039: Transient Modeling of Integrated Nuclear Energy Systems with Thermal Energy Storage and Component Aging and Preliminary Model Validation via Experiment 16-026: Computationally Efficient Prediction of Containment Thermal Hydraulics using Multi-Scale Simulation: Feasibility Study	Focus on transient modeling of integrated, hybrid energy systems that would incorporate nuclear and renewable generators, electricity production, an industrial process, and thermal energy storage. Develop a technical basis for coarse-grained computational fluid dynamics capability that is needed for high-fidelity analysis of containment thermal-hydraulics.



University	Projects	Scope
Ohio State University	<p>14-009: Development of an INL Capability for High-Temperature Flow, Heat Transfer, and Thermal Energy Storage with Applications in Advanced Small Modular Reactors, High-Temperature Heat Exchangers, Hybrid Energy Systems, and Dynamic Grid Energy Storage Concepts</p> <p>15-039: Transient Modeling of Integrated Nuclear Energy Systems with Thermal Energy Storage and Component Aging and Preliminary Model Validation via Experiment</p> <p>15-141: Interfacing MOOSE Components to Enhance Capability</p>	Design, development, and analysis of effective and robust high-temperature heat transport systems and working fluids to support successful deployment of advanced high-temperature reactor systems for both power generation and non-electric applications. Transient modeling of integrated, hybrid energy systems that would incorporate nuclear and renewable generators, electricity production, an industrial process, and thermal energy storage. Develop a non-equilibrium chemical model for assessment of chemical and corrosion reactions among coolant, cladding, and fuel. It also includes the assessment of hydrogen production and cladding oxidation.
Oregon State University	<p>15-039: Transient Modeling of Integrated Nuclear Energy Systems with Thermal Energy Storage and Component Aging and Preliminary Model Validation via Experiment</p> <p>15-060: Development of Efficient TREAT Modeling Capabilities with Graphite Data Improvement</p>	Focus on transient modeling of integrated, hybrid energy systems that would incorporate nuclear and renewable generators, electricity production, an industrial process, and thermal energy storage. Research efforts to support modeling and experimentation related to the transient test reactor.
Pennsylvania State University	16-046: Development of a Synergistic Approach to Study Irradiated Materials Using Coupled Experiments and Simulation	Demonstrate use and integration of MOOSE/BISON/MARMOT capabilities to shorten the nuclear fuel development cycle.
Saint Louis University	15-146: Tailoring the Kinetic Function of a Surface through Electronic Effects of Nanoscale Architecture	Focus of industrial processes to build chemical intermediates from more abundant shale gas resources, focusing on selective dehydrogenation of light alkanes.
Texas Tech University	16-081: Modeling Thermite Reactions	Develop a model that emulates thermite reactions using the MOOSE framework.

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University	Projects	Scope
University of California, Berkeley	16-071: Evaluation of Advanced Digital Neutron Imaging Systems for PIE of Nuclear Fuel	Test advanced digital neutron imaging detectors for their applicability to evaluate irradiated nuclear fuel.
University of California, Davis	15-023: Development of Stochastic Three-Dimensional Soil Response Capability in MOOSE to Provide Design and Beyond Design Basis Seismic Motions for Nuclear Facilities	Develop two new numerical capabilities in the MOOSE-based application, MASTODON: (1) three-dimensional seismic wave propagation in time domain computations and (2) stochastic finite elements (allows for direct computation of variability in soil material properties).
University of California, Irvine	16-098: Nuclear Nonproliferation Applications of $^{14}\text{C}$ Analyses by Accelerator Mass Spectrometry	Development of a gas phase $^{14}\text{C}$ sample preparation manifold system for ultra-trace $^{14}\text{C}$ accelerator mass spectrometry analysis.
University of Connecticut	14-079: Second Generation Switchable Polarity Solvent Draw Solutes for Forward Osmosis	Explore membrane/ switchable polarity solvent compatibility addressing water flux data, reverse solute flux, and solute/membrane parameters for several membranes.
University of Idaho	14-010: Use of Linear Variable Differential Transformer-Based Methods to Detect Real-Time Geometry Changes During Irradiation Testing 14-093: All Hazards Critical Infrastructure Knowledge Framework 15-144: Investigation of Sonication-Assisted Electrolytic Reduction of Used Oxide Fuel in Molten Salt 16-002: Advanced Carbon Feedstock Processing Using Ionic Liquids 16-085: Production of Fluoroanion Targets for Accelerator Mass Spectrometry 16-096: Supporting Operator Performance and Situation Awareness in Highly Automated Nuclear Power Plants 16-176: Development of Direct Carbon Fuel Cells	Develop in-pile detection of changes in geometry to understand real-time behavior during irradiation testing of fuels and materials in high flux U.S. Material and Test Reactors. Develop advanced knowledge discovery and decision support methodologies to improve the nation's capability to conduct infrastructure interdependency analysis in order to better prepare for, protect against, respond to, recover from, and mitigate all hazards. Investigate the feasibility of improving electrolytic reduction of used oxide fuel in molten salts using ultrasound techniques; Develop efficient coal and biomass conversion

University	Projects	Scope
	16-187: Micro-Scale Technique to Evaluate Grain Boundary Cohesion of Irradiated Alloys	technologies. Develop the technology for rapid production of sample targets for actinide analysis using AMS through the use of actinide fluoroanion salts produced using novel fluorinating ionic liquid and extracted using supercritical CO <sub>2</sub> ; investigate the impact of high levels of automation in complex, safety critical systems, and to demonstrate ways of enabling optimal situation awareness for human operators in those systems. Develop robust, reduced-temperature direct carbon fuel cells. Develop a technique to quantify grain boundary cohesion and its impact on fracture behavior in irradiated alloys, by utilizing transmission electron microscopic in situ cantilever testing in concert with multi-scale modeling;
University of Louisiana, Lafayette	16-096: Supporting Operator Performance and Situation Awareness in Highly Automated Nuclear Power Plants	Investigate the impact of high levels of automation in complex, safety critical systems and to demonstrate ways to enable optimal situation awareness for human operators in those systems
University of Michigan	14-045: End-To-End Radiation Detector Enhancements for Improved Safety and Security in Safeguarded Facilities	Demonstrate that radiation detection systems, such as portal monitors, located at safeguarded facilities can be significantly enhanced through an end-to-end improvement of the scintillation-based detection equipment used within these systems.
University of Montana	16-003: Recycling of Tantalum-Containing Waste Materials to Recover Tantalum Metal	Develop a high-temperature electrochemical process with a view to reclaiming tantalum metal from its primary and secondary resources.

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University	Projects	Scope
University of New Mexico	<p>14-075: Development of Tools and Methodologies for Uncertainty Quantification and Validation for Multiphysics Fuel Performance Simulation</p> <p>15-039: Transient Modeling of Integrated Nuclear Energy Systems with Thermal Energy Storage and Component Aging and Preliminary Model Validation via Experiment</p> <p>15-060: Development of Efficient TREAT Modeling Capabilities with Graphite Data Improvement</p> <p>15-094: Evaluation and Demonstration of the Integration of Safeguards, Safety, and Security by Design</p> <p>15-142: New In-Core Neutron Diagnostics</p>	<p>Develop a mathematical and software framework to perform uncertainty propagation for multi-physics simulations involving both neutronics and fuel performance simulations. Focus on transient modeling of integrated, hybrid energy systems that would incorporate nuclear and renewable generators, electricity production, an industrial process, and thermal energy storage. Modeling and experimentation related to the transient test reactor. Provide nuclear reactor operators and designers with a cost-effective method of integrating safety, security, and safeguards considerations into design of the reactor and provide a risk-informed decision-making platform for safety, security, and safeguards personnel. Demonstrate the use of optical method for live-time monitoring of reactor neutron flux.</p>
University of Utah	<p>16-152: Wireless Radio Frequency Signal Identification and Protocol Reverse Engineering</p>	<p>Build a tool, wireless radiofrequency signal Identification and protocol reverse engineering that is usable in an operational context for automatically analyzing the radiofrequency emanations and wireless protocols from a black box device and performing automated wireless protocol reverse engineering.</p>
University of Wisconsin	<p>14-098: Irradiation Effects in Uranium Dioxide</p>	<p>Use both in situ and ex situ transmission electron microscopy observation of defect nucleation and evolution under ion irradiation to understand the radiation damage mechanisms in <math>\text{UO}_2</math>.</p>

University	Projects	Scope
University of Wyoming	15-146: Tailoring the Kinetic Function of a Surface through Electronic Effects of Nanoscale Architecture 16-002: Advanced Carbon Feedstock Processing Using Ionic Liquids 16-176: Development of Direct Carbon Fuel Cells 16-187: Micro-Scale Technique to Evaluate Grain Boundary Cohesion of Irradiated Alloys	Focus on industrial processes to build chemical intermediates from more abundant shale gas resources. Develop efficient coal and biomass conversion technologies. Develop robust, reduced-temperature direct carbon fuel cell. Develop a technique to quantify grain boundary cohesion and its impact on fracture behavior in irradiated alloys by utilizing transmission electron microscopic situ cantilever testing in concert with multi-scale modeling.
Washington State University	15-135: Dynamic Simulations for Large-Scale Electric Power Networks in Real-Time Environment using Multiple RTDS	Test and evaluate necessary components for geographically distributed real-time simulation using real-time simulators and interconnectivity between different locations.

### Industry Collaborations

Industry	Project	Scope
Idaho Power Company	14-086: Development of a Microgrid/Smartgrid Testbed for ESL and Super Lab Initiative, with Load Variability Characterization and Control for Renewable Energy Integration	Focus on understanding, testing, and modeling the system dynamics of real-world microgrids and distributed energy resources at multiple scales and geographic locations to successfully increase renewable penetration.
LUNA Innovations	16-149: In-Core Qualification of Developmental Instrumentation	Perform ex-core characterization of several sensors under development for in-core use.
NuScale	16-096: Supporting Operator Performance and Situation Awareness in Highly Automated Nuclear Power Plants	Investigate the impact of high levels of automation in complex, safety critical systems and to demonstrate ways of enabling optimal situational awareness for human operators in those systems.

## 2016 Project Summaries — Appendices

Industry	Project	Scope
ON Semiconductor	14-086: Development of a Microgrid/Smartgrid Testbed for ESL and Super Lab Initiative, with Load Variability Characterization and Control for Renewable Energy Integration	Focus on understanding, testing, and modeling the system dynamics of real-world microgrids and distributed energy resources at multiple scales and geographic locations to successfully increase renewable penetration.
PH.D.s Co.	14-037: Advanced Nuclear Material Characterization Technology for Security Applications	Explore new sensor combinations that can be used in support of national objectives for detecting nuclear and radiological materials
POWER Engineers	14-086: Development of a Microgrid/Smartgrid Testbed for ESL and Super Lab Initiative, with Load Variability Characterization and Control for Renewable Energy Integration	Focus on understanding, testing, and modeling the system dynamics of real-world microgrids and distributed energy resources at multiple scales and geographic locations to successfully increase renewable penetration.
RTDS	15-135: Dynamic Simulations for Large-Scale Electric Power Networks in Real-Time Environment using Multiple RTDS	Test and evaluate necessary components for geographically distributed real-time simulation using real-time simulators and interconnectivity between different locations.
Technology Insights	14-009: Development of an INL Capability for High-Temperature Flow, Heat Transfer, and Thermal Energy Storage with Applications in Advanced Small Modular Reactors, High-Temperature Heat Exchangers, Hybrid Energy Systems, and Dynamic Grid Energy Storage Concepts	Focus on design, development, and analysis of effective and robust high-temperature heat transport systems and working fluids to support successful deployment of advanced high-temperature reactor systems for both power generation and non-electric applications.
USAF Cyber Security Fellows Program	14-032: Remote Vulnerability Analysis of CAN Bus Networks	Discover new technologies capable of enhancing the cyber security of the CAN bus networks that are used within a typical passenger vehicle, with applications that can also address transportation systems related to aerospace, rail, and maritime.

**National Lab Collaborations**

National Laboratory	Projects	Scope
Argonne National Laboratory	16-055: Capability Extension for Multiscale, Multi-Application development within the Multiphysics Object-Oriented Simulation Environment	Develop and implement several experimental algorithms within the framework, enabling forward-thinking analysis and one-of-a-kind capabilities for challenging problems facing the numerical simulation and modeling community.
Institute for Energy Technology/Halden Reactor Project	14-010: Use of Linear Variable Differential Transformer-Based Methods to Detect Real-Time Geometry Changes During Irradiation Testing	Develop in-pile detection of changes in geometry to understand real-time behavior during irradiation testing of fuels and materials in high flux U.S. materials test reactors.
Los Alamos National Laboratory	16-010: Development of a Fully Coupled Radiation Damage Production and Evolution Simulation Capability	Couple simulation of radiation damage (MyTRIM), microstructure evolution (MARMOT), nuclide transmutation, and neutronics (MAMMOTH) to help spatially resolve damage calculation capability with ballistic mixing informed by MyTRIM, transmutation in MARMOT informed by MAMMOTH, and residual defect densities implemented using the phase field formalism.
National Physical Laboratory	16-149: In-Core Qualification of Developmental Instrumentation	Perform ex-core characterization of several sensors under development for in-core use.
National Renewable Energy Laboratory	15-135: Dynamic Simulations for Large-Scale Electric Power Networks in Real-Time Environment using Multiple RTDS	Test and evaluate necessary components for geographically distributed real-time simulation using real-time simulators and interconnectivity between different locations.
The French Alternative Energies and Atomic Energy Commission	16-149: In-Core Qualification of Developmental Instrumentation	Perform ex-core characterization of several sensors under development for in-core use.



## **Appendix D**

### **List of Interns and Postdocs**



## Appendix D

### List of Interns and Postdocs

#### 2016 LDRD Interns

Name	Affiliation	LDRD Project No.
Adams, Ben	Oregon State University	15-039, 16-050
Aggarwal, Rachit	Oregon State University	15-039, 15-141
Alberti, Anthony	Ohio State University	15-060
Allen, Jared C	Brigham Young University - Idaho	16-002
Argyle, Jordan M	Brigham Young University - Provo	16-013
Baltaji, Omar	University of Illinois - Urbana Champaign	15-023
Bao, Han	North Carolina State University	16-026
Baset, Aniqua Z.	University of Utah	16-152
Becker, Christopher D.	University of Utah	16-152
Bratton, Ryan	Texas Tech University	16-081
Burns, Allison	Oregon State University	16-050
Castellano, Andrea	Politecnico di Milano	15-023
Chatterjee, Payel	North Carolina State University	16-040
Chavez, Cynthia	University of Pittsburgh	15-111
Chen, Minghui	Ohio State University	14-009
Chorney, Maureen P.	Montana Technical University	16-003
Colby Sorrell, Nina	North Carolina State University	15-060
Daniels, M. Tucker	North Carolina State University	15-039
Dustin, Jeremiah	University of Idaho	15-144
Drew, Keith	Idaho State University	15-100
Ellis, Matt	Massachusetts Institute of Technology	15-060
Fathi, Nima	University of New Mexico	15-039
Frick, Konor	North Carolina State University	15-039
Fullarton, Michele	University of Florida	14-026
Ghassemi, Pedram	North Carolina State University	16-010
Giegel, Sam	Idaho State University	16-070
Goldstein, Michael	University of Pittsburgh	15-111
Green, Jason P.	Texas Technical University	16-081
Grinder, Mara	Michigan State University	14-045
Guo, Qingti	Ohio State University	15-039
Harter, Jackson	Oregon State University	15-060

<b>Name</b>	<b>Affiliation</b>	<b>LDRD Project No.</b>
Haugen, Carl	Massachusetts Institute of Technology	15-060
Hazelip, Naomi	Boise State University	15-100
Hembree, Harvey R.	Portland State University	15-083
Hill, Rachael A.	Brigham Young University - Idaho	16-096, 15-111
Hix, Jay D.	Eastern Idaho Technical College	14-037
Holschuh, Thomas	Oregon State University	14-037
Hoover, Kyle	Oregon State University	15-039
Hrisko, Joshua	Pennsylvania State University	15-040
Hu, Guojun	University of Illinois - Urbana Champaign	16-017
Hurley, Bridger	Montana Tech	16-003
Jambrina Gomez, Ana	Pennsylvania State University	14-075
Jarrold, Patrick	Oregon State University	16-050
Jin, Miaomiao	Massachusetts Institute of Technology	16-058
Kersten, Bethany R.	University of Idaho	14-095
Kim, Hun	Ohio State University	14-009
Koonce, Tony	Idaho State University	15-013
Kursteiner, Karen	California State University - San Bernardino	16-106
Kuznicki, Andrew	Weber State University	16-152
Ladd, Jacob C.	Brigham Young University - Provo	16-017
Lambert, Janine N.	University of Florida	15-094
Lee, Jieun	University of Idaho	15-144
Li, Huijuan	Ohio State University	15-039, 15-141
Lim, Ken	University of CA, Berkeley	15-146
Lin, Alex	Carnegie Mellon University	15-111
Lin, Linyu	North Carolina State University	16-026
Liu, Cheng	Ohio State University	15-039
Lyle, Brendan	University of Pittsburgh	15-111
McAlister, Justin D.	California Polytechnic University	14-025
McClusky, Andrew	University of Pittsburgh	15-111
Merryman, Bobbi	University of New Mexico	15-094
Mikkelsen, Daniel	University of North Carolina	15-039
Misenheimer, Corey	North Carolina State University	15-039
Morrell, Jonathan	Massachusetts Institute of Technology	16-036
Morris, Joe	University of New Mexico	15-142
Moseley, Maxwell C.	Mississippi State University	14-093
Nichels, John	North Carolina State University	15-039
Numanoglu, Ozgun	University of Illinois - Urbana Champaign	15-023

<b>Name</b>	<b>Affiliation</b>	<b>LDRD Project No.</b>
Odeniyi, Adefolaoluwami	North Carolina State University	15-039
Oh, Seungje	Ulsan National Institute of Sci. & Tech, Korea	15-144
Osorio Ramirez, Julian D.	Florida State University	15-135
Osterhause, Nicholas	University of New Mexico	15-094
Pelka, Sara	University of New Mexico	15-142
Petti, Andrew N.	University of Wisconsin	14-093
Pharr, Alexander	North Carolina State University	15-040
Pietrykowski, Michael	Ohio State University	15-039, 15-141
Pizzocri, Davide	Politecnico di Milano	14-031
Pollock, Kevin	University of Pittsburgh	15-111
Rashidnia, Bobak	Ohio State University	15-039, 15-141
Reece, Christian	Cardiff University	15-146
Reimund, Kevin	University of Connecticut	14-079
Repukaiti, Reyixiati	Oregon State University	16-050
Ryals, Matthew	University of New Mexico	15-060
Schoonen, Alexander L.	Montana State University	15-125
Shy, Daniel		14-045
Singhal, Anant	North Carolina State University	15-060
Song, Eric	Ulsan National Institute of Sci. & Tech, Korea	15-144
Sosa, Charles S	University of Michigan	14-037, 14-045
Stack, Daniel	Massachusetts Institute of Technology	15-039
Tan, Shuai	University of Wyoming	15-146, 16-002
Teter, Marco	Oregon State University	15-039
Teter, Lucas	Oregon State University	15-039, 16-050
Tu, Lei	University of Idaho	16-017
Umbrage, Brittany L.	University of New Mexico	16-040
Waliga, Abby	University of Pittsburgh	15-111
Weaver, Gabriel	University of Illinois at Urbana, Champaign	16-106
Wegman, Kevin	Ohio State University	14-009
Weitzel, David B.	University of New Mexico	14-075, 15-039
Westmen, Bjorn	Oregon State University	15-039
Wormald, Jonathan	North Carolina State University	15-060
Wu, Huarui	Massachusetts Institute of Technology	16-036
Xue, Sha	Ohio State University	15-141
Zhang, Xiaoqin	Ohio State University	14-009
Zhu, Muzhi	Ohio State University	15-039
Zhu, Yangmo	North Carolina State University	16-017, 16-026

**2016 LDRD Postdocs**

Postdoc Employee	University	LDRD Project
Abir, Muhammad	Missouri University of Science and Technology	16-036
Adhikari, Birendra	University of Colorado	14-079
Antonio, Daniel J.	University of Nevada	15-032
Baroi, Chinmoy	University of Saskatchewan	16P6-002FP
Calderoni, Patrick	University of California	16-149
Diao, Weijian	University of South Carolina	16P6-002FP
Diaz Aldana, Luis A	Ohio University	15-140
Fullerton, Michele	University of Florida	14-026
Heathman, Colt R	Washington State University	15-002
Jiang, Wen	Duke University	14-031, 16-187
LaBrier, Daniel	University of New Mexico	15-060
Liu, Maolong	University of New Mexico	15-060
Luo, Yusheng	Florida State University	15-135
Lwin, Soe	Lehigh University	16P6-002FP
McNally, Josh	Boston University	14-079
Meher, Subhashish	University of North Texas-Denton	14-078
Pawar, Gorakh M	University of Utah	15-128
Price, Patrick	Boise State University	15-128
Rai, Durgesh	Massachusetts Institute of Technology	16-036
Redekop, Evgeniy	University of Oslo, Norway	15-146
Shi, Shanbin	Ohio State University	15-141
Short, Michael	Massachusetts Institute of Technology	16-058
Shrestha, Keshav	University of Houston	15-032
Skifton, Richard	University of Idaho	14-010, 16-149
Veeraraghavan, Swetha	California Institute of Technology	15-023
Wang, Congjian	Purdue University	14-075
Weaver, Gabriel	University of Illinois, Urbana-Champaign	16-106
Yadav, Vaibhav	University of Iowa	16-040
Yoon, Su Jong	Seoul University	14-009
Zhang, Wei	Michigan Technological University	16-096
Zhou, Jing	University of Utah	15-128, 16-081

## **Appendix E**

### **Patents**





## Appendix E

### Patents

#### Patents LDRD Program 2016

Project No.	Patent No.	Title	Inventors	Date Granted	CC	Description
99-292	9,428,401	Dry Recycle of Spent Nuclear Fuel – Separation of the Rare-Earth Fission Product Poisons	James W. Sterbentz and Jerry D. Christian	8/30/16	Nuclear and Radio Chemistry	Various methods are available for recycling used nuclear fuel in order to recover more of the energy potential of a given quantity of fuel. Aqueous and electrometallurgical methods are well known, but a dry recycling method exists that does not extract plutonium, use toxic chemicals, or produce large volumes of waste. This patent describes a method for improving dry recycling further by providing a way to separate neutron-absorbing fission product poisons – primarily rare-earths – from the used nuclear fuel. Removal of the rare-earth neutron poisons reduces the amount of enriched uranium oxide feed material needed to re-use the fuel, thus improving the efficiency of this used fuel recycling method.
GB103	9,234,228	Alteration of Enzyme Stability and Activity via Covalent Modification	David N. Thompson, William A. Apel, Vicki S. Thompson, David William Reed, and Jeffrey A. Lacey	1/12/16	Biological and Bioprocess Engineering Chemical and Molecular Science	It is recognized that bacteria glycosylate (attach a carbohydrate) their proteins. Glycosylation of a protein has been shown to assist in the protein's stability and activity, modulate physical properties (such as solubility), and protect against its destruction. The glycosylated enzymes from <i>Alicyclobacillus acidocaldarius</i> have enhanced activities under industrially relevant conditions such as with acid and at very high temperatures. This patent describes some of the genes responsible for glycosylation of enzymes in the thermoacidophilic microorganism <i>Alicyclobacillus acidocaldarius</i> .
13-093	9,215,587	TT: Self-Generating Fault Tolerant Encryption Key	Hussein Moradi, Behrouz Farhang, and Rangam Subramanian	12/15/15	Advanced Computer Science, Visualization, and Data	In wireless radio communication, spread-spectrum techniques involve increasing a signal's bandwidth for a variety of reasons, including to avoid jamming and reducing the probability of detection or interception. However, secure communication may be compromised if an eavesdropper has already obtained the spreading code being used for transmission of a spread-spectrum signal. This invention advances

Project No.	Patent No.	Title	Inventors	Date Granted	CC	Description
						deployment of spread-spectrum systems by dynamically selecting the spreading code used by a pair of communicating devices in such a way that it will be inaccessible to an eavesdropper. The channel reciprocity between the communicating parties is taken advantage of to set up a common secret key (a spreading gain) among them. The self-generated key remains unknown to an eavesdropper whose different physical location, with respect to the legitimate parties, leads to a different set of channel parameters.
14-079	9,399,194	TT: Thermolytic Draw Solute for Osmotically Driven Membrane Processes	Aaron D. Wilson and Christopher J. Orme	7/26/16	Chemical and Molecular Science	Decreasing water supplies throughout much of the industrialized world have necessitated new methods and systems for utilizing water that contains contaminants or impurities. Additionally, certain industries have a need for safer, more energy-efficient methods and systems for removing water from a target material or solute. Traditional purification methods include thermal flash evaporation and reverse osmosis (a process where water is separated from solutes through a membrane by application of pressure). Forward osmosis circumvents several of the deficiencies of reverse osmosis by using a draw solution with a greater osmotic pressure than the feed liquid. The concentrated solution pulls the water through the membrane, leaving the contaminants behind, and the water can be extracted from the solution with low-grade heat. This patent relates to methods and draw solutions for treating a liquid using an N-cycloalkyl-cycloalkylamine as a switchable polarity solvent.

## 2016 Project Summaries — Appendices

Project No.	Patent No.	Title	Inventors	Date Granted	CC	Description
10-075	9,369,866	Methods and Apparatuses Using Filter Banks for Multi-Carrier Spread Spectrum Signals	Hussein Moradi, Behrouz Farhang, and Carl A. Kutsche	6/14/16	Advanced Computer Science, Visualization, and Data	Wireless services in harsh and hostile environments must overcome challenges such as intentional jamming (e.g., brought about by an enemy during wartime) and unintentional interference (e.g., collision with the presence of other devices due to limited spectral resources). Known for their robustness, spread spectrum systems extend information across more bandwidth than is required by the data rate. Multi-carrier spread spectrum is a particular form of spread-spectrum designed to be resistant to narrow or partial band interference. This invention uses filter-bank techniques to generate and detect multi-carrier spread-spectrum signals that can carry information at a very low power level distributed over the frequency spectrum. The signal transmission is kept at or below the noise level of other signals, leading to a low probability of detection and interception.

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