Nuclear Process Science Initiative NEWSLETTER

Pacific Northwest NATIONAL LABORATORY

The Nuclear Process Science Initiative (NPSI) is a Pacific Northwest National Laboratory (PNNL) internally-funded effort to advance nuclear process science capabilities to meet national needs in environmental management, nonproliferation, and other areas. The five-year initiative was launched in mid-2015.

***NPSI's vision** is to understand, harness, and exploit interfacial phenomena controlling the behavior of materials in nuclear processing.*

Researchers are working in three "thrust" areas:

***Science Thrust 1**:*
Legacy Waste

***Science Thrust 2**:*
Nuclear Security

***Science Thrust 3**:*
Analytical Capabilities

NPSI Leadership:

April 2019

Reid's Notes

NPSI progress sparks questions about the future

By Reid Peterson, Initiative Lead

It has been a busy past few months to say the least. We have completed a couple projects and started new ones.
With each successful endeavor (we've wrapped eight projects to date, transitioned a couple others to new efforts, and will complete three projects this fiscal year), it prompts the question of "what's next?" for NPSI.

Answering that will be our ongoing focus as the initiative moves toward conclusion in 2020. With input from our advisory committee and PNNL leadership, we have been giving serious thought to how we will not only successfully conclude the initiative, but position our outcomes for broader impact at PNNL, for follow-on funding from clients, and for perhaps even new research efforts. We're excited about the possibilities. Stay tuned.

Achieving Initiative Milestones

Initiative Lead: Reid Peterson
Deputy Lead: Brienne Seiner
Thrust 1 Lead: Reid Peterson
Thrust 2 Lead: Jon Schwantes
Thrust 3 Lead: Kevin Rosso
Project Coordinator: Barb Beller
Finance: Debbie Lucas
Communications: Tim Ledbetter

Visit the NPSI Website
Contact Us

NPSI-hosted Seminar:

Dr. Ian Farnan Speaks April 1 at PNNL

Dr. Ian Farnan of the Department of Earth Sciences at the University of Cambridge, UK, is scheduled to speak at PNNL on "Model Approaches to Spent Nuclear Fuel Durability." This seminar, hosted by NPSI, will be held Monday, April 1, 11 a.m., in the EMSL Auditorium. All interested staff are welcome.

Dr. Farnan's research focuses on an atomistic understanding of radiation damage and aqueous corrosion processes in nuclear materials, including nuclear waste forms and their natural analogues, fuels, and clads.

Journal Bestows "HOT" Status on Paper

Physical Chemistry Chemical Physics (PCCP) has selected a

As I noted, we launched two new projects early this fiscal year:

- "Particle Size and Density Measurement," a one-year effort focused on developing a new measurement method with potential applications for radioactive waste slurries. **Philip Gauglitz** is the principal investigator.

- "Characterization of Carbonaceous Surface Deposits on Irradiated Iron Aluminide ($FeAl_3$) Coatings." An eight-month effort, this project is led by **Walter Luscher** and seeks to deliver new insights into characterization of irradiated materials.

Our teams also recently completed two projects:

- "Ion Implantation and Characterization of Epsilon Metal Phase Formation in Ceria," led by **Ram Devanathan** (see the cerium oxide article below to learn about one of this project's accomplishments).

- "Fundamental Insights into Gamma-Radiation Effects at Complex Oxide-Water Interfaces from First Principles Simulations," led by **Sebastien Kerisit**.

Both of these three-year efforts delivered solid outcomes, and we're exploring opportunities for follow-on funding to further apply and advance resulting capabilities.

In January, we updated the NPSI advisory committee on our projects and many other aspects of the initiative during a mid-year review meeting. We received the committee's endorsement for our current direction, as well as suggestions to enhance efforts going forward. We'll provide the committee with a more comprehensive accounting of NPSI progress during our Annual Review, set for July 9-10.

Reid

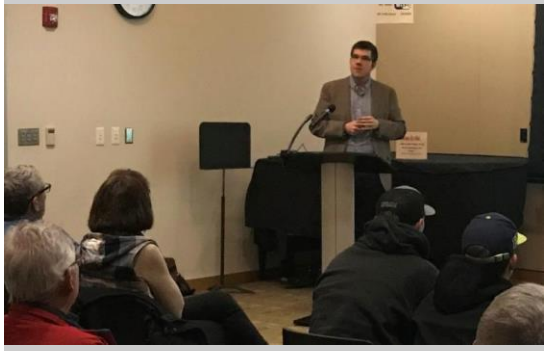
NPSI-funded paper as a HOT manuscript for 2018.

"Surfactant-enhanced heterogeneity of the aqueous interface drives water extraction into organic solvents," was authored by Michael Servis and Aurora Clark of Washington State University (WSU). Clark serves in a joint appointment with PNNL.

The research was funded as part of NPSI's project, "Interfacial Diffusion and CRUD Formation at the Liquid-Liquid Interface of Solvent Extraction Processes Research," led by **Amanda Casella**.

The HOT designation indicates the handling editor or referees viewed the paper as among "the hottest work published in *PCCP*" in 2018.

NPSI Project Lead Gives Talk at PNNL Seminar Series Event



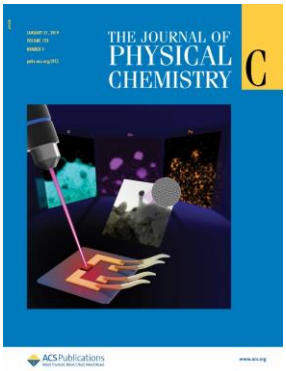
Steven Spurgeon, principal investigator of the NPSI project, "Damage Mechanisms and Defect Formation in Irradiated Model

Study of Doped Cerium Oxide Sheds Light on Metallic Nanoparticles

Novel particle inquiry and PNNL artwork are featured on journal cover

There's much to learn about what happens when fission is occurring inside nuclear fuel. NPSI researchers are providing some new insights into that process--without exposing themselves to the challenge or dangers of working with radioactive materials.

The researchers used a non-radioactive surrogate material, cerium oxide, that's very similar to the uranium oxide fuel used in nuclear reactors. The objective was to understand and predict the precipitation (formation) and growth of metallic particles in irradiated nuclear fuel using the surrogate doped with five metals. The team employed advanced microscopes to characterize particle precipitation *in situ* during thermal treatment and ion irradiation. Read a summary of the project, methods, and results.



The research and its outcomes are chronicled in the paper, "In Situ Study of Particle Precipitation in Metal-Doped CeO_2 during Thermal Treatment and Ion Irradiation for Emulation of Irradiating Fuels," published in the January 25, 2019, edition of *The Journal of Physical Chemistry C*.

Cover art representing the work was created by PNNL's Nathan Johnson, a graphic designer in the Communications and Information Technology Directorate.

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Concluding Thrust 2 Projects Provide Insights into Epsilon

Systems," spoke at the PNNL Community Science and Technology Seminar Series on February 19 in the Richland (WA) Public Library.

His topic, "Modern Day Alchemy: Building the Future Atom by Atom," provided a look at how contemporary ideas and tools are leading to new materials and other advances.

He spoke on the same topic on February 22 in Richland for a WSU Tri-Cities' Civil and Environmental Engineering Seminar.

Researchers Recognized with "Of-the-Year" Publication Award

At a special event in December, PNNL's Energy and Environment Directorate (EED) honored the team that authored a NPSI-funded review paper.

The team received the "Publication of the Year" award at the annual EED Of-the-Year ceremony, which honored various research and project accomplishments. The paper, "Review of the Scientific Understanding of Radioactive Waste at the U.S. Department of Energy Site," was published in *Environmental Science and Technology*.

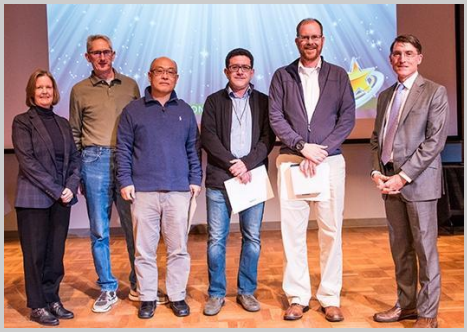
Particles, Nucleation, and Materials Precipitation

A number of NPSI projects concluded in fiscal year 2018. The NPSI Newsletter is reviewing some of the outcomes of these projects. The three featured below are associated with Science Thrust 2 (Nuclear Security). Additional Thrust 2 projects that have been completed will be highlighted in the next edition.

Project: **Modeling the Interfacial Effects, Partitioning, and Production Routes of Epsilon Particles in Uranium Oxide**
Principal Investigator: **Richard Clark**
Outcomes: The project used molecular scale analyses and first principles modeling to study formation and properties of noble metal phase particles. These particles, referred to as epsilon phase or five metal phase (based upon historic descriptions of their composition), include molybdenum, technetium, ruthenium, rhodium and palladium. They form during irradiation of nuclear fuel and are the source of deleterious effects related to fuel integrity loss over time. As part of the project, researchers re-established a Radiochemical Processing Laboratory (RPL) capability that enabled a full cross-section of an irradiated uranium dioxide fuel pellet to be prepared and removed from hot cells. Other experimental work, which involved collaboration with the University of Missouri, provided the first viability of microscale irradiations.

The project enabled important discoveries related to particle formation and behavior, including: 1) the existence of tellurium, the "sixth" element, in noble metal phase particles; 2) close association of the noble metal phase with important fission products, including xenon; 3) identification of noble metal phase particles and xenon within the outer zirconium fuel cladding; 4) elucidation of noble metal phase formation and behavior that implicates these species in the local production of high-pressure fission gas bubbles in fuel, ultimately leading to cladding corrosion at the fuel/cladding interface. These discoveries offer far-reaching implications for next generation fuel designs.

Project: Fundamental Understanding of Nucleation Processes to Assess Solution Stability and Phase Growth and Genesis
Principal Investigator: **Gregg Lumetta**



EED Associate Laboratory Director Jud Virden (far right) and EED Chief Science and Technology Officer Sue Clark (far left) recognize the authors, from left, ***Gregg Lumetta****,* ***Jaehun Chun****,* ***Eugene Ilton****, and* ***Reid Peterson****. Not shown:* ***Edgar Buck*** *and* ***Richard Daniel****.*

Peterson Joins Publication's Editorial Board

NPSI's **Reid Peterson** began a three-year term on the editorial board of *Environmental Progress and Sustainable Energy (EP&SE)* in January. He was invited to serve by the publication's editor, Martin Abraham of Youngstown State University.

Peterson will work with the board to address policy questions, assist with manuscript reviews, and participate in the group's annual meeting.

The publication, issued quarterly by the American Institute of Chemical Engineers, covers a wide range of environmental issues, including waste treatment and remediation, air pollution, sustainability, and sustainable energy.

Outcomes: The focus was on creating a physicochemical framework for examining and predicting precipitation reactions relevant to nuclear materials processing and nuclear forensics. Experimental and computational methods probed plutonium oxalate nucleation and growth (europium oxalate was used in most experiments). Various microscopy tools were employed, investigating growth rates down to nanometer resolution. Growth rates were fit to theoretical models to deduce growth pathways. The project explored a novel approach for developing electronic parameters to describe crystallinity and surface tension of nanocrystals (using sodium chloride as a case study).

Other project outcomes included creation of a Cryo-Transmission Electron Microscopy capability in PNNL's Radiochemical Processing Laboratory, and successful modeling of europium oxalate growth with a diffusion limited growth mechanism, which found that localized inhomogeneities due to mixing played an important role in the crystal growth. The project also applied machine learning to predict surface tensions controlling kinetics of nucleation and crystallization processes.

Project: Phase Field Modeling of Microstructure Development in Plutonium (IV) Oxalate Precipitation
Principal Investigator: David Abrecht
Outcomes: More information is needed about microstructural evolution of precipitating solid materials--including the ways crystal shapes and structures form--to improve nuclear forensics capabilities. The focus of the project was on development of phase field models (in which the solid-liquid interface is treated as a continuous mathematical field) aimed at capturing, in plutonium oxalate, the correlation between the precipitating conditions and final structures, and then translating this information into a predictive capability.

The project's progress included development of the physical constants for the plutonium oxalate system needed to describe crystallization behavior in phase field models, and establishment of thermodynamic properties of the solutions from which plutonium oxalate is normally precipitated. In fiscal year 2018, the project's results gained the attention of, and funding from, the Department of Homeland Security, and the project was transitioned out of NPSI.

NPSI Publications

October 2018 - March 2019

Devanathan, R., W. Jiang, M.A. Conroy, K. Kruska, T.C. Droubay, J.M. Schwantes. 2019. **"Hexagonal Close-packed High Entropy Alloy Formation under Extreme Processing Conditions,"** Journal of Materials Research, 34(5): 709-719. DOI: 10.1557/jmr.2018.438. Invited paper.

Jiang, W., M.A. Conroy, K. Kruska, M.J. Olszta, T.C. Droubay, J.M. Schwantes, C.A. Taylor, P.M. Price, K. Hattar, R. Devanathan. 2019.**"In situ Study of Particle Precipitation in Metal-doped CeO2 during Thermal Treatment and Ion Irradiation for Emulation of Irradiating Fuels,"** *Journal of Physical Chemistry C,* 123:2591-2601. **DOI:** 10.1021/acs.jpcc.8b11027.

Shen, Z., E.S. Ilton, M.P. Prange, C.J. Mundy, S.N. Kerisit. 2018. **"Diffusion Mechanisms of Radiolytic Species in Irradiated Al (Oxy-) Hydroxides,"** *Journal of Physical Chemistry C,* 122(50): 28990-28997. **DOI:**10.1021/acs.jpcc.8b07809.

Servis, M.J., A.E. Clark. 2019. **"Surfactant-enhanced Heterogeneity of the Aqueous Interface Drives Water Extraction into Organic Solvents,"** *Physical Chemistry Chemical Physics,* 21(6): 2866-2874. **DOI:**10.1039/c8cp06450d. NPSI Project Principal Investigator: Amanda Casella.

PNNL-SA-142383

Spurgeon Organizing Second Electron Microscopy Workshop

NPSI's **Steven Spurgeon** is putting together a workshop in August at the Microscopy Society of America (MSA) annual Microscopy and Microanalysis Conference in Portland, OR. The workshop's focus will be on the future of electron microscopy.

MSA approached Spurgeon about organizing the workshop as a pre-meeting congress, which occurs prior to the main conference, after learning about a similar event he organized at PNNL last fall. That meeting, the inaugural Next-Generation Transmission Electron Microscopy (NexTEM) workshop, was held October 8-10 in Discovery Hall.

While the NexTEM event at PNNL was not sponsored by NPSI, some of the meeting's topical areas aligned with the initiative's research activities.

In NPSI, Spurgeon leads a project focused on high-resolution studies of radiation damage in oxides.