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

October 2018

Reid's Notes

Laser focus on publications is bringing prominence to our work

By Reid Peterson, Initiative Lead

From the beginning, publishing has been a key NPSI emphasis.

If we're not sharing the results of our research, then obviously we're not contributing knowledge to the broader science community, nor meeting the expectations of PNNL and the Laboratory Directed Research and Development program that funds NPSI's efforts. As we work to not just meet but exceed expectations, our publication emphasis is on achieving a healthy balance of quality placements versus number of placements, and I think we've been successful in this regard.

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

NPSI Leadership:

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

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NPSI Researchers Anticipate Future Collaborations from Denver Meeting


Sweet

Luke Sweet, who leads the NPSI project, "Characterization of Radiation Induced Materials Defects Across Scales," co-presented a workshop at the prestigious Denver X-ray Conference in Colorado in August.


Corbey

NPSI colleague **Jordan Corbey** also attended, presenting a poster.

"The workshop was titled 'Line Profile Analysis,' and we had good attendance, with probably 50 to

For the recently-completed fiscal year 2018, NPSI researchers made significant progress against goals. Twelve journal articles--an increase of four over last fiscal year--were published in peer-reviewed publications. Some recent examples include papers in the *Journal of Nuclear Materials*, *Computational Materials Science Journal* and *Journal of Chemical Physics*. One of our projects earned the cover of *Analytical Chemistry*, and you can read more about that in a separate article in this newsletter.

We recently recognized several of our principal investigators and, by extension, their project teams, for publication accomplishments in fiscal year 2018. **Jaehun Chun**, **Sebastien Kerisit**, **Dallas Reilly**, and **Edgar Buck** each had two papers published during the year.

Of course, the goal is exponential progress, which should be achievable considering our increasing number of project accomplishments and discoveries. NPSI researchers are working on multiple papers that are in various stages of preparation and submission, so I'm very confident we'll continue the upward trend in the final two years of the initiative.

Reid

New Microscope Reveals Xenon in Fuel Cladding

Identification of the chemical element delivers new knowledge and demonstrates benefit from application of an advanced microscopy capability

In a pioneering discovery, NPSI researchers, using a new instrument at the Radiochemical Processing Laboratory (RPL), have detected and imaged xenon in a section of cladding, or the thick outer layer of a fuel rod.

The revelation, the first observation of its kind, will provide insights into how the fission product got into the cladding and chemically fractionated during nuclear reactor operations. Such information could lead to advances in future fuel designs.

75 people," Sweet explains. The workshop was organized by Matteo Leoni from the University of Trento, Italy, who presented along with Sweet and two others--Jim Cline and Marcus Mendenhall, both from the National Institute of Standards and Technology. The workshop provided a historical overview of diffraction line profile analysis, assessments of the state of the art, and some practical examples and advice.

Sweet indicates the workshop, along with Corbey's poster and a separate presentation he gave, generated a number of conversations with conference attendees interested in the science being conducted. "We ended up starting the seeds of several new collaborations and I am very excited to see where things go," Sweet says.

Corbey's poster was titled, "Structural Investigation of Plutonium Oxalate Species and Comparison of Their Oxide Products." Sweet's second presentation was, "X-ray Line Profile Analysis of Plutonium Oxide."

Paper and Graphic Earn Journal Cover

A NPSI-funded paper and image were featured on the cover of the July 17, 2018, edition of *Analytical Chemistry*.

The paper, "Micro-Raman Technology to Interrogate Two-Phase Extraction on a Microfluidic

To obtain the result, researchers introduced the radioactive fuel sample into the GrandARM scanning transmission electron microscope, installed at RPL just this year and one of only three such instruments in the U.S. The microscope uses a 300-kiloelectron volt beam, which enables the exploration of thicker regions of a specimen. The instrument's probe aberration correction helps maintain excellent spatial resolution, and the dual energy-dispersive detectors, unique to the GrandARM, allow rapid detection of xenon and other minor species.

Xenon is the most common fission product generated in spent nuclear fuel. The first observations of the element were made in uranium dioxide fuels. Attempts to locate xenon in cladding failed previously because instruments with lower-energy beams could not penetrate the regions of the sample where the gas was intact.

The GrandARM resides in RPL's new Quiet Suite for high-end microscopy, and is one of four microscopes expected to revolutionize PNNL's micro-analytical characterization capabilities for studying radiological materials.

The RPL is a Hazard Category II Non-Reactor Nuclear Facility. Along with other PNNL assets, the laboratory provides key state-of-the-art nuclear research resources of value to the nation.

Concluding Thrust 1 Projects Provide Insights into Colloidal Interactions, Filtration

Several NPSI projects concluded in fiscal year 2018. The NPSI Newsletter will review some of the outcomes of the projects, starting with two from Science Thrust 1 (Legacy Waste) focused on colloidal interactions and filtration, respectively.

Project: Correlation of Colloidal Interactions and Macroscopic Rheology in Concentrated Electrolyte Solutions
Principal Investigator: Jaehun Chun
Outcomes: This project has provided a scientific knowledge base to accomplish safe and efficient nuclear waste treatment processing, in addition to proper design and operation of waste disposition processes. The project created a customized atomic force microscopy/dynamic force spectroscopy capability for direct measurement of

Device," results from a collaboration between PNNL, the College of Idaho, Washington State University (WSU), and Spectra Solutions, a Small Business Innovation Research program partner.

The paper was authored by PNNL's **Susan Asmussen** and **Amanda Lines**, and visiting collaborator Gilbert Nelson from the College of Idaho. Co-authors were PNNL's **Amanda Casella**, **Danny Bottenus**, **Sam Bryan** and **Sue Clark**, who serves in a joint appointment with WSU.

PNNL graphic designer Rose Perry provided the cover image, which represents the authors working in a "lab-on-a-chip" to perform in-situ Raman spectroscopy and chemometric modeling of a biphasic nuclear chemical separation, demonstrating a successful application of in-situ process monitoring within a microfluidic channel.

This methodology enables work with small sample volumes and obviates the need for further chemical treatment, both great benefits when conducting solvent extraction in the harsh environments associated with nuclear fuel cycle separations.

boehmite particles, as well as a particle-based simulation framework to predict bulk suspension rheology. Combined with a project-developed in situ scanning electron microscope capability for particle aggregation/identification, collectively these advances provide basic understanding of how particle interactions are correlated to long-range ordering of colloidal waste particles and bulk rheology in tank-waste-relevant systems.

Project: Particle-Filter Surface Interactions and Dynamics in the Presence of Cross-Flow
Principal Investigator: Richard Daniel
Outcomes: With a goal of providing capabilities to enhance effectiveness and reduce time and cost associated with Hanford tank waste filtration, the project made progress on multiple fronts. Early on, a Filter Visualization Cell was designed and built for direct imaging of particle accumulations on filter elements. This, along with the project team's creation of chemically-representative waste simulants, made it possible to evaluate, at the bench-scale, crossflow and dead-end filter system performance. In the final year of the project, models were developed that offer new knowledge about gel-polarization in crossflow filters and fouling in dead-end filters--advances that help provide a predictive capability for filter performance and effectiveness. One of the project's additional benefits was a new collaborative relationship with Howard University.

NPSI Progress Gains Advisory Committee Endorsement

Following the NPSI Annual Review on July 11-12, the initiative's Advisory Committee concluded NPSI is on track to make solid contributions to PNNL's nuclear science and technology capabilities.

The committee, made up of internal and external experts, determined the work conducted in NPSI's three thrust areas--Legacy Waste, Nuclear Security, and Analytical Capabilities--is relevant to initiative objectives and beneficial for sustaining PNNL's Radiochemical Processing Laboratory (RPL).

"The committee was very pleased with the progress that we have made over the past year and is looking forward to

This image appeared on the cover of Analytical Chemistry, illustrating the "lab-on-a-chip" concept.

Spurgeon Helps Shape Future of TEM

NPSI Principal Investigator **Steven Spurgeon** participated in a five-day workshop, "Development of an Integrated Transmission Electron Microscope," in Telluride, CO, June 17-21.

Steven, representing PNNL, was one of two dozen microscopy leaders from national laboratories and academia who gathered with instrument manufacturers to completely reimagine the design of the transmission electron microscope (TEM).

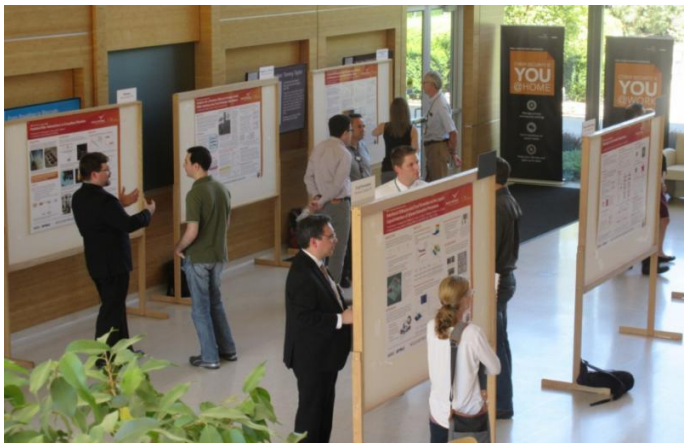
"Our goal is to develop an instrument that will grant us access to unprecedented spatial, chemical, and temporal dimensions," explains Spurgeon. "We're entering a new frontier of electron microscopy that will enable us to solve grand challenges in nuclear science,

seeing what more we can accomplish in the last two years of the initiative," says NPSI Lead **Reid Peterson**.

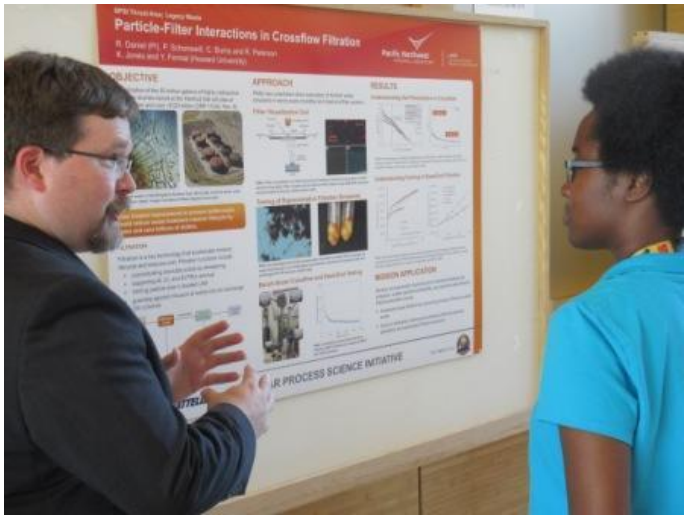
During the review, Reid and other NPSI leaders provided updates on progress in the initiative's thrust areas. The meeting also included presentations about NPSI's support to PNNL and Energy and Environment Directorate objectives, and the status of funding, staffing, and equipment issues that potentially impact initiative operations.

The Advisory Committee toured the RPL to gain a better understanding of the facility, particularly new instrumentation and equipment.

Scenes from the NPSI Annual Review Poster Session on July 11:



The poster session provided project staff with the opportunity to highlight the objectives and accomplishments of their research.



*Filtration project Principal Investigator **Richard Daniel** (left) explains some of his team's work to a poster session attendee.*

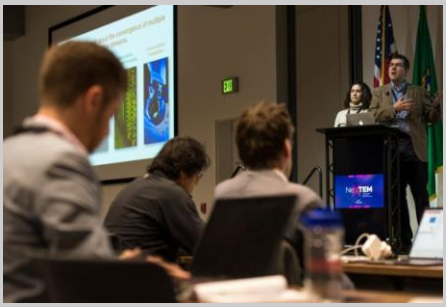
quantum computing, and energy storage."

Workshop participants presented ideas from their current research, identified limitations of existing microscopes, and shared their vision for the microscope of tomorrow. Spurgeon highlighted work from his NPSI project focused on high-resolution studies of irradiated thin-film oxides, and also discussed microscopy investments that are positioning PNNL as a hub for cutting-edge electron microscopy research.

He is helping to author a review article for a journal that will summarize the workshop's conclusions.

The Next Step in TEM: NexTEM Workshop

Spurgeon also helped coordinate the workshop, "Next Generation Transmission Electron Microscopy (NexTEM)," October 8-10, at PNNL. Meeting outcomes will be reported in the next edition of the NPSI Newsletter.



The NexTEM Workshop was held at PNNL, October 8-10.

From left, ***Susan Asmussen*** *and* ***Amanda Casella*** *discuss their project poster with Christopher Ramos from DOE's National Nuclear Security Administration. Ramos participated in the Annual Review as a guest Advisory Committee member. Amanda and Susan's poster received the "Best Poster" award based on reviewer feedback.*

Shawn Riechers *(back to camera) discusses the iRAD-AFM capability with visitors to his poster.*

NPSI Publications

April 2018 - October 2018

Reilly, D.D., M.T. Athon, J.F. Corbey, I.I. Leavy, K.M. McCoy, J.M. Schwantes. 2018. "**Trace Element Migration During UF4 Bomb Reduction: Implications to Metal Fuel Production, Worker Health and Safety, and Nuclear Forensics**," Journal of Nuclear Materials, 510:156-162.
DOI: 10.1016/j.jnucmat.2018.07.052.

Garrett, K.E., A.M. Ritzmann, F.N. Smith, S.H. Kessler, R. Devanathan, J.J. Henson, D. Abrecht. 2018. "**First Principles Investigation of the Structural and Bonding Properties of**

Hydrated Actinide (IV) Oxalates, An(C2O4)2.6H2O (An = U, Pu)," Computational Materials Science Journal, 153:146-152. DOI: 10.1016/j.commatsci.2018.06.033.

Prange, M.P., X. Zhang, E.S. Ilton, L. Kovarik, M.H. Engelhard, S.N. Kerisit. 2018. "**Electronic Response of Aluminum-Bearing Minerals**," Journal of Chemical Physics, 149(2): 024502. DOI: 10.1063/1.5037104.

Buck, E.C., R.S. Wittman, C.Z. Soderquist, B.K. McNamara. 2018. "**Monitoring Bromide Effect on Radiolytic Yields using in-situ Observations of Uranyl Oxide Precipitation in the Electron Microscope**," RSC Advances, 8:18227-18233. DOI: 10.1039/c8ra01706a.

Nelson, G.L., S.E. Asmussen, A.M. Lines, A.J. Casella, D.R. Bottenus, S.B. Clark, S.A. Bryan. 2018. "**Micro-Raman Technology to Interrogate Two-Phase Extraction on a Microfluidic Device**," Analytical Chemistry. DOI: 10.1021/acs.analchem.7b04330. **(featured on cover of journal)**.

Prange, M.P., X. Zhang, M.E. Bowden, Z. Shen, E.S. Ilton, S.N. Kerisit. 2018. "**Predicting Surface Energies and Particle Morphologies of Boehmite (γ¬AlOOH) from Density Functional Theory**," Journal of Physical Chemistry C. 122(19): 10400-10412. DOI:10.1021/acs.jpcc.8b00278.

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Soltis J.A., W.C. Isley, M. Conroy, S.M. Kathmann, E.C. Buck, G.J. Lumetta. 2018. "**In situ Microscopy Across Scales for the Characterization of Crystal Growth Mechanisms: The Case of Europium Oxalate**," CrystEngComm. 20:2822-2833. DOI: 10.1039/C7CE01450C.

PNNL-SA-139102