Advances in pH and phosphate monitoring enhance safety in nuclear fuel recycling

October 28 2021, by Kimberlee Papich

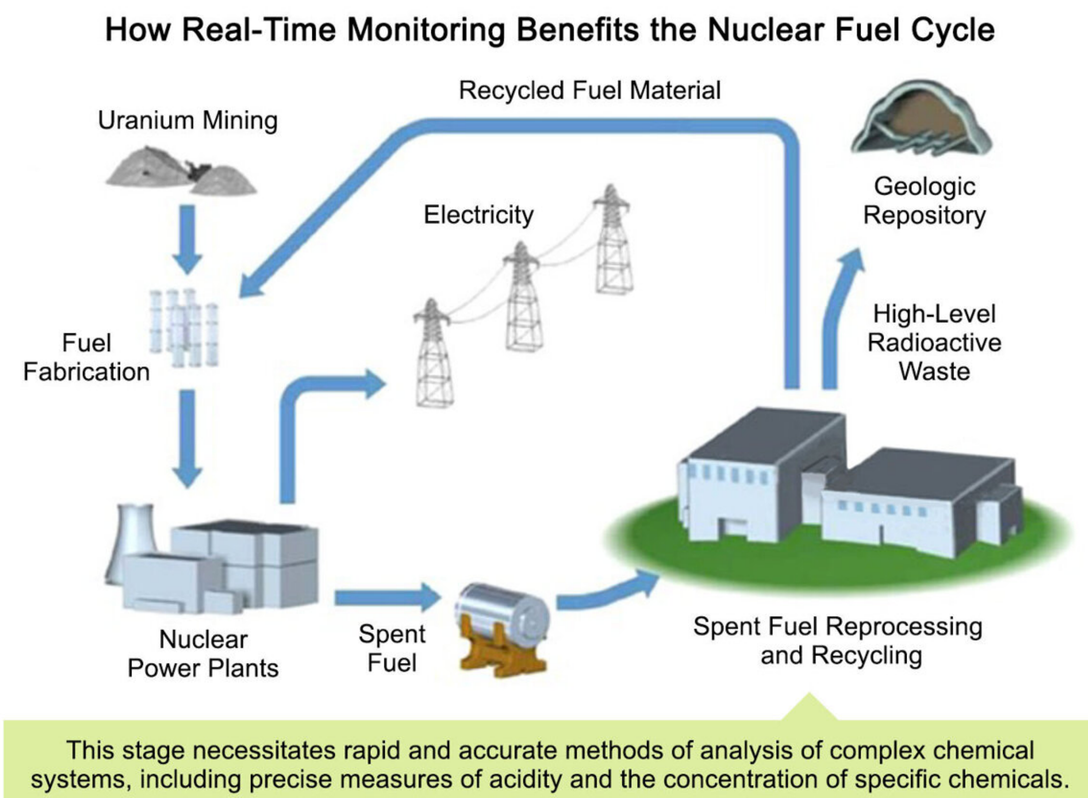


Illustration shows where real-time, online monitoring and accurate measurement of chemical species can positively affect the nuclear fuel cycle. Credit: Nathan Johnson | Pacific Northwest National Laboratory

Picture a scenario in which a highly toxic substance, like dissolved nuclear fuel, is being recycled in a reprocessing plant. Until now, workers inside the plant had very limited information about what's in the solution to inform next steps. But that's changing, thanks to the innovative work led by two interns at Pacific Northwest National Laboratory (PNNL). Now, without opening, sampling, or otherwise handling the substance, they developed an easy way to determine its acidity and chemical components—using a method as simple as taking a picture. The approach allows researchers to understand the composition of the solution safely and effectively and to quickly decide a path forward.

Researchers are keenly interested in finding a means to better determine the pH level and chemical structure of dissolved nuclear fuel—after it's been fired in a reactor—to improve the safety and efficiency of reprocessing and enhance the overall sustainability of the nuclear fuel cycle. As the fuel is reprocessed to separate reusable fuel material from radioactive waste, variables like alkalinity and other chemical involvement can interfere with how well materials can be recovered and recycled. Experts are also looking to increase the safety of workers and the environment by reducing contact with radioactive waste, with potential for these methods to extend into many other industries and quality control processes.

Enter PNNL and its decades of experience in extracting resources from spent nuclear fuel, and its pioneering work into remote testing and online monitoring. This is the expertise informing the contactless technique the two PNNL interns recently adapted to better measure the acidity of highly radioactive solutions, as well as the concentration of specific chemicals.

Located in Richland, Washington, adjacent to the Department of Energy's (DOE's) Hanford Site, PNNL matched its mentors to students and used funding from DOE's Office of Nuclear Energy to expand the

Lab's real-time testing and continuous monitoring capabilities to help industry partners like DOE with more efficient and safer reprocessing and recycling of spent nuclear fuel. And because successful fuel reprocessing can reduce the burden of radioactive waste disposal, real-time monitoring also helps address critical environmental and national security needs.

As a result, PNNL's reimagining of pH measurement and chemical analysis is not only gaining attention for the Laboratory itself, but also for the two interns who researched this issue.

Leading the charge

Hope Lackey and Andrew Clifford are those two interns. Lackey first heard about PNNL while working toward her undergraduate degree in environmental studies at the College of Idaho. Her chemistry professor and mentor, Gilbert Nelson, whom she credits with sparking her love for chemistry, encouraged Lackey to take part with him in PNNL's Visiting Faculty Program in 2017. Her pH measurement and chemical analysis research occurred during her Science Undergraduate Laboratory Internships (SULI) experience at PNNL in 2018. Clifford, also a SULI intern mentored by Nelson at the College of Idaho, partnered with Lackey between his junior and senior year, while studying for a dual bachelor's in chemistry and math/physics.

Their innovative work, under the mentorship of Sam Bryan, a PNNL Lab fellow and chemist who leads PNNL's efforts in real-time testing and continuous monitoring, was featured in two recent *Analytical Chemistry* journal articles. In "Reimagining pH Measurement: Utilizing Raman Spectroscopy for Enhanced Accuracy in Phosphoric Acid Systems," Lackey and her co-authors delve into complex chemical systems like Hanford tank waste and radioactive solutions.

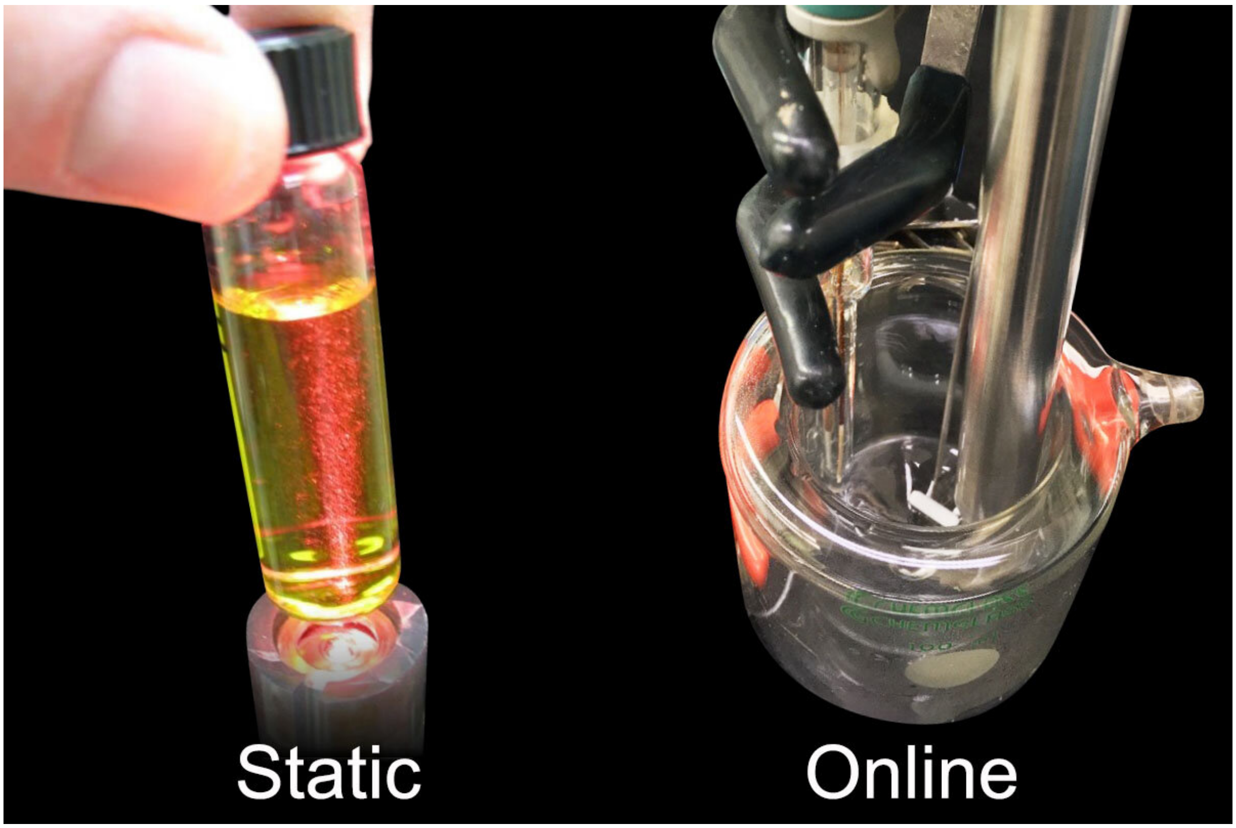
Radioactive tank waste is a byproduct of nuclear fuel, which is used to power more than 440 commercial nuclear reactors globally, including 93 in the United States. Spent nuclear fuel cannot stay long-term in sites' aging tanks, meaning experts must dispose of it or reprocess it. While the United States currently plans on disposing of its spent nuclear fuel, it is also conducting research and development to advance reprocessing technologies that could be deployed in a nuclear fuel cycle. PNNL, a key player in this research and development (R&D), focuses on improving real-time operation of reprocessing. This research necessitates rapid and accurate methods of analysis of complex chemical systems. But there are several difficulties associated with the traditional method of determining acidity, called pH monitoring.

Problems with traditional pH monitoring

Offline lab analyses, with the use of traditional pH probes, to process the tank waste can be costly and time-consuming. The process also requires a worker to go into the process stream and manually collect a sample to determine which chemicals need to be added, to confirm if the radioactive materials have left solution, and more. Manual collection exposes the worker to a potentially very hazardous solution.

The sample then only represents the solution at the point in time it was retrieved, offering limited accuracy in dynamic solution environments like those associated with nuclear fuel recycling. Additionally, numerous challenges are inherent to the probes themselves, including their fragility and requirement for optimal conditions, limiting their usefulness in real-time monitoring applications. This motivated Lackey and the team from PNNL's Radiochemical Processing Laboratory, one of the nation's premier—and enduring—resources for applied nuclear science and technology, to look to remote, immediate, and online analyses of waste acidity.

At the core of the team's remote pH sensing technique is measuring light's interaction with chemical bonds, or optical monitoring of visible light spectra, using an analytical technique called Raman spectroscopy. In contrast to traditional probes, Raman probes are physically robust and can function for extended periods in harsh environments.



In the data collection phase of her research, PNNL intern Hope Lackey measured pH in both static (offline) and online solutions. Lackey was able to show speciation in the online spectra, despite its background contents. Credit: Pacific Northwest National Laboratory

The approach also employs a branch of artificial intelligence and computer science known as machine learning. This type of learning, called chemometrics, creates a set of rules (an algorithm) for a computer to follow in its calculations to turn spectral response into a measure of acidity. In turn, sample analysis occurs in real time, with more rapid and cost-effective results. This online monitoring also precludes the worker from coming into contact with the solution.

"Basically, instead of manually dealing with these caustic solutions, we're adapting robust probes to shine an intense light on solutions, but the 'camera' we use doesn't make colored images, it gives us 'pictures' in real time to record the solution's response to light," said Lackey.

The novelty of this approach earned Lackey a DOE Innovations in Nuclear Technology R&D Award from the Office of Nuclear Energy, which recognized the technique's inherent benefits for nuclear waste processing and worker and environmental safety.

As their mentor, Bryan is uniquely positioned to comment on the interns' innovations.

Said Bryan of Lackey and her achievements, "Hope is really doing transformational science here. She is mimicking existing probes by building tools that can adapt to extremely harsh environments to generate the type of easy-to-understand information researchers and operators need for fundamental studies and to design new recycling schemes."

Identifying chemicals present in complex solutions

The second journal article, which was given cover status, was "Raman Spectroscopy Coupled with Chemometric Analysis for Speciation and Quantitative Analysis of Aqueous Phosphoric Acid Systems." Co-lead authored by Clifford and Lackey, the paper describes an additional application for online, optical monitoring—to characterize and quantify, in real time, chemicals present in complex solutions like radioactive waste.

This approach builds off the techniques of the first article and uses chemometrics to measure a new parameter in the samples—the concentrations of chemicals, in this case, phosphates. Under different levels of acidity, phosphate can take four chemical forms based on proton removal. Clifford and Lackey's technique quantifies each type of phosphate and the total phosphate, at any pH.

Online monitoring for phosphates allows nuclear experts to perform initial separations of the chemical, which is crucial to assure phosphates do not interfere with overall processing. Similarly, this detection may be of use in the analysis of other types of phosphates present during waste purification and storage.

Said Clifford, "material tracking and accountability are really important in these nuclear processes. What we're offering with online monitoring is smoother operation by enabling real-time analysis and modification of system processes."

Although the two papers cover potential benefits specific to nuclear waste processing, Amanda Lines, who was previously mentored by Bryan and is now his colleague in the Radiochemical Processing Laboratory, is quick to point out the additional possibilities inherent to Lackey and Clifford's findings.

"These approaches align well with PNNL's mission around energy and environmental cleanup and help us maintain our position as leaders in online monitoring research, but what I find compelling are the possibilities to improve how other chemical processes are deployed, outside of nuclear," Lines said.

"Many commercial and industrial processes requiring quality control would benefit from quick and easy pH measurement and phosphoric acid detection—things like fertilizers and pharmaceutical drugs," commented Lines. "Regardless of application, these interns have made the ease of process monitoring conducive to optimization."

More information: Hope E. Lackey et al, Reimagining pH Measurement: Utilizing Raman Spectroscopy for Enhanced Accuracy in Phosphoric Acid Systems, *Analytical Chemistry* (2020). DOI: 10.1021/acs.analchem.9b05708

Andrew J. Clifford et al, Raman Spectroscopy Coupled with Chemometric Analysis for Speciation and Quantitative Analysis of Aqueous Phosphoric Acid Systems, *Analytical Chemistry* (2021). DOI: 10.1021/acs.analchem.1c00244

Provided by Pacific Northwest National Laboratory

Citation: Advances in pH and phosphate monitoring enhance safety in nuclear fuel recycling (2021, October 28) retrieved 5 July 2024 from https://phys.org/news/2021-10-advances-ph-phosphate-safety-nuclear.html