Benchmark data set validates global nuclear reactor codes

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Oak Ridge National Laboratory researchers re-evaluated used nuclear fuel rods from a commercial reactor and reduced data uncertainties by an order of magnitude compared with previous measurements taken at a different lab. ORNL's more accurate data enabled validation of nuclear simulation codes and explanation of anomalies. Credit: Oak Ridge National Laboratory, U.S. Dept. of Energy

Nearly 100 commercial nuclear reactors supply one-fifth of America's energy. For each fuel rod in a reactor assembly, only 5 percent of its energy is consumed before fission can no longer be sustained efficiently for power production and the fuel assembly must be replaced. Power plants currently store the used fuel on-site. Information on the composition of the used fuel is essential for the design of safe storage, transportation and final repository facilities and for inspection and verification to safeguard nuclear materials. Improved accuracy in prediction of the spent fuel isotopic composition leads to increased efficiency in the facility designs and higher confidence in the safeguard protocols.

To demonstrate the accuracy of computer codes for predicting the isotopic composition of used nuclear fuel and other radioactive waste, researchers in the United States have conducted experimental studies aimed at accurately characterizing used nuclear fuel to ensure it can be stored safely and efficiently. Using radiochemical techniques, they have analyzed the isotopic contents of fuel rods from many commercial nuclear reactors and compared the results with calculated values from nuclear reactor fuel simulation codes. One of the cornerstone validation data sets includes fuel from the Three Mile Island Unit 1 (TMI-1) reactor that was measured between 1998 and 2000.

In 2012, Catherine Romano of the Department of Energy's Oak Ridge National Laboratory (ORNL) identified a need to better understand the relationship between the isotopic content of used fuel as measured using nondestructive analysis (NDA) and the actual isotopic content of the fuel to strengthen nuclear safeguards. Romano is a member of ORNL's Nuclear Material Detection and Characterization group and principal investigator of a National Nuclear Security Administration project through the Office of Defense Nuclear Nonproliferation to develop NDA methods for used fuel. In 2013, members of the project team at ORNL re-analyzed two fuel rods from the TMI-1 reactor that were previously used in a cornerstone data set. They used NDA measurements (from neutron and gamma ray detectors) and improved destructive radiochemical assay methods (of samples from dissolved fuel rods) to provide very accurate correlations between the NDA signals and measured isotopic concentrations. In addition, they compared the measured concentrations to simulations and cross-checked results against the earlier measurements.

Published in the Annals of Nuclear Energy, the re-analysis used improved radiochemical methods developed at ORNL by Joe Giaquinto with Jeffrey Delashmitt and Tamara Haverlock of ORNL's Chemical Sciences Division and characterized more than 50 different isotopes and 16 elements in the fuel rods with high accuracy. It produced an experimental data set with uncertainties many times smaller than those obtained by the earlier radiochemical analysis. With this improved standard of experimental accuracy, modeling and simulation experts at ORNL—first author Ian Gauld, Jianwei Hu and Germina Ilas, all of the Reactor and Nuclear Systems Division—applied the radiochemical analysis and NDA data to validate codes widely used by the nuclear safeguards research community.

Improving experimental accuracy

"We created a benchmark for other nuclear analytical chemistry labs to follow for detailed used fuel characterizations," said Giaquinto, who led the chemical analytics of the new study. "We can provide experimental data with a much improved degree of confidence that modelers can use to evaluate predicted results from their computational models. This gives them a more accurate data set when validating reactor codes based on the empirical data. We improved the experimental end of the validation process."

Giaquinto and colleagues procured the key lanthanide standards that made the high-precision measurements possible from the National Isotope Development Center (NIDC), whose Business Office is located at ORNL. The NIDC, which is supported by the DOE Isotope Program (managed by the Office of Science), coordinates isotope production and sales across DOE Isotope Program–funded facilities. Giaquinto's team purchased additional standards from a commercial vendor.

Combining high-pressure, high-resolution separation techniques with isotopically enriched standards, Giaquinto, Delashmitt and Haverlock were able to rapidly isolate multiple elements in sets of actinides and lanthanides. The elementally pure fractions were then analyzed using isotope dilution, the most accurate technique available for quantification with mass spectrometry.

"For isotope dilution, a well certified enriched isotope of an element is added gravimetrically to the working solution, or spiked, before chemical processing," Giaquinto explained. "The enriched isotope is now the calibrant and is equilibrated into the system with the unknowns. From there on out, whatever you do to that sample, the calibrant follows the same chemistry process as the unknown, allowing for a high degree of precision in the measurement compared to an external calibrant."

That technique produced uncertainties of 1.5 to 2 percent, a dramatic improvement over uncertainties of 10 percent or more produced with previous techniques.

The scope of nuclear resources at ORNL made the achievement possible. "We have a facility where they could handle the fuel," Giaquinto said. "We have hot cells where we could do detailed dissolution. And we have the breadth of nuclear expertise from reactor modeling to NDA to analytical chemistry."

Capturing complexities using modeling and

simulation codes

Simulation codes are used to model the complexities of a reactor and track the creation and destruction of more than 1,600 nuclides in nuclear fuel during its life in the reactor. The reactor core contains more than a hundred fuel assemblies, each in turn filled with hundreds of zirconium-alloy-clad uranium oxide rods that heat water to generate power. Each fuel rod experiences a different history in the reactor resulting from its unique position in the fuel assembly.

Researchers widely use the SCALE nuclear systems modeling and simulation code package, developed at ORNL, to model fuel assembly designs and complex reactor operating conditions and calculate changes in the isotopic content of nuclear fuel as it is burned. Using ORNL's new measurement data for the TMI-1 fuel samples, the current study yielded excellent agreement between code predictions and measurements and provided more accurate inventory data for comparison with NDA safeguards measurements. Moreover, the new high-precision measurements have resolved discrepancies between previous measurements and simulated values for some isotopes in one of the TMI-1 cornerstone data sets that have perplexed the scientific community for more than 10 years.

Gauld's code-validation work is part of a global effort to compare simulation results against data from experiments conducted internationally. Representing DOE, Gauld chairs the Organisation for Economic Co-operation and Development's Nuclear Energy Agency subgroup on assay data for used nuclear fuel, which is compiling a public database of measurements taken on more than 600 fuel samples. The information from the ORNL study will be entered into that database. "We want to capture all measurements of this type that have been done in the world for different reactor types and make it a central, go-to database for experiment used fuel inventory data," Gauld said. The database will provide not only measurement data, but also fuel design information and all primary references needed for modeling. It will reside both at the NEA Data Bank in Paris and at the Radiation Safety Information Computational Center in Oak Ridge.

More information: I.C. Gauld et al. Re-evaluation of spent nuclear fuel assay data for the Three Mile Island unit 1 reactor and application to code validation, *Annals of Nuclear Energy* (2016). DOI: 10.1016/j.anucene.2015.08.026

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