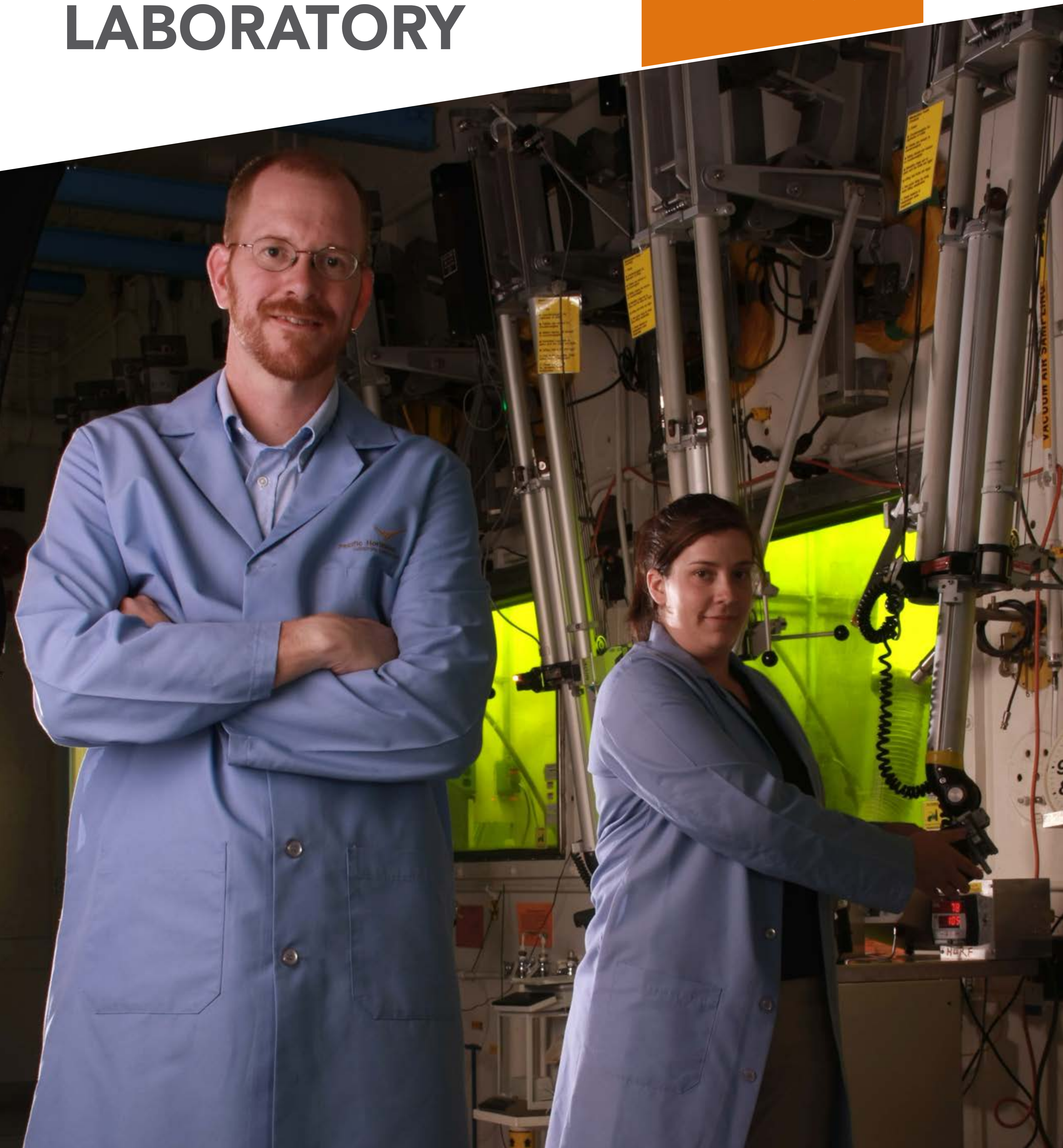
RADIOCHEMICAL PROCESSING LABORATORY

Pacific Northwest
NATIONAL LABORATORY



Located on the U.S. Department of Energy Hanford Site in south-central Washington State, the Radiochemical Processing Laboratory is an essential facility at the Pacific Northwest National Laboratory that supports enduring missions in environmental management, nuclear energy, national security, and science.

Radiochemical Processing Laboratory

Across the nation, solutions are needed to clean up radiological and hazardous wastes, safeguard America against nuclear proliferation, ensure safe operation of nuclear power plants, and process and dispose of nuclear fuel used to generate electricity.

The Radiochemical Processing Laboratory (RPL), a Hazard Category II Non-Reactor Nuclear Facility in Richland, Washington, provides scientific discovery and technology solutions for these challenges. RPL is one of the nation's premier—and enduring—resources for applied nuclear science and technology. Stewarded by the U.S. Department of Energy, this facility serves other federal agencies, including the Department of Homeland Security and Department of Defense. In all cases, RPL offers one-of-a-kind capabilities for developing and deploying innovative radiological material processes and solutions that enable America to overcome its energy, environment, economic, and security challenges.

RPL's unique laboratories and instrumentation are underpinned by a team of nationally-distinguished researchers who are in the vanguard of nuclear science and technology and are credited with novel, groundbreaking discoveries and applied solutions.

In support of PNNL's objective to disrupt nuclear trafficking, scientists at the RPL use chemical characterization methods to perform nondestructive analysis of radiological materials.

Radiochemistry and Processing

Radiochemical Process Development at All Scales

Our primary expertise lies in development, scale-up, and deployment of first-of-a-kind processes to solve problems in environmental remediation, nuclear fuel-cycle, and national security applications. These proficiencies include extensive experience with DOE high-level tank waste and actinide process streams.

Among the key features of the RPL are extensive specialized facilities and instrumentation to identify and quantify chemical species and radioactive isotopes in simple and complex media. Our scientists and engineers work with materials ranging from highly radioactive samples to highly dispersible isotopes to trace levels of radionuclides. The RPL is one of only two facilities within the U.S. Department of Energy (DOE) complex capable of broad-spectrum R&D on large quantities of fissile materials.

Our capabilities in radiochemical process science and engineering can be applied to

- developing new radiochemical separation methods to support advanced fuel cycles and environmental remediation
- researching, testing, and validating process flowsheets
- designing, installing, and optimizing radiochemical process systems
- devising engineered systems for processing toxic and highly radioactive materials
- discovering nuclear process and material signatures.

Our gloveboxes and remote manipulation facilities allow us to design, install, and test small-scale radiochemical processes to support the development of large-scale counterpart facilities, such as the Hanford Tank Waste Treatment and Immobilization Plant for the vitrification of Hanford tank waste contents. At the RPL, actual tank waste samples have been analyzed to determine the fluid-flow properties of the complex high-salt, solids-laden waste. Nonradiological waste simulants that possess similar fluid-flow properties have been developed at the RPL to support large-scale mixing and transport tests.

RPL glovebox capabilities support U.S. Department of Homeland Security (DHS) missions in nuclear forensics. The DHS' National Technical Nuclear Forensics Center has been developing a plutonium (Pu) material database to support the attribution of interdicted nuclear materials. In an effort to understand the effects of process variations on the characteristics of the finished Pu product, researchers in the RPL have previously duplicated the Bismuth Phosphate process and are currently investigating the Plutonium Uranium Redox Extraction (PUREX) processes at the 200g Pu scale.

Centrifugal contactor bank installed in a shielded glovebox for separations flowsheet testing with transuranic materials.

Expert Chemical and Physical Separations

The RPL's unique facilities and multidisciplinary staff enable separations research that extends from the molecular level conceptualization to the testing of prototype flowsheets for industrial applications. Our expertise in the fundamental chemistry of radionuclides and our knowledge of radiochemical separations technologies enable us to develop innovative solutions to meet our sponsors' diverse needs.

The focus of separations research in the RPL is on research and development with the actual radioactive materials. Capabilities include ion exchange, solvent extraction, electrochemical membrane separation, and cross-flow ultrafiltration techniques. We perform specialized flowsheet testing using 2-cm centrifugal contactors for solvent extraction, ion exchange columns for resin-based separations, and a cell-unit filter for radioactive and nonradioactive cross-flow ultrafiltration. In addition, we employ spectroscopic techniques, calorimetric methods, and other equipment to determine the properties of the materials to be processed, both chemical (thermodynamics, actinide speciation) and physical (rheology, thermal conductivity, particle-size distribution). The resulting information provides us with the insight necessary to design better separations, or to refine and optimize chemical separation processes.

Separations research in the RPL also encompasses developing new sample processing applications that lead to dramatic increases in productivity in radioanalytical laboratories. Examples include development of new separations media and the design and testing of automated radiochemical separations for trace-level to high-level chemical analysis.

Research in the volatilization of fission products and actinides is opening new doors for the processing of nuclear materials, with applications to national security and fundamental science.

One of the unique capabilities of the RPL is actinide research. Here, a researcher works with a plutonium sample using one of several gloveboxes in the RPL.

Tritium Processing

The RPL offers capabilities for testing tritium processing flowsheets and providing direct design input or solutions for full-scale facilities such as the U.S. Department of Energy Tritium Extraction Facility. Capabilities include:

- Determining processing parameters needed to meet facility functional design criteria
- Determining input for process flowsheets
- Supplying input on equipment design or specifications
- Providing input for process model development.

The laboratories at the RPL have high-temperature (1100°C) vacuum furnaces for tritium extraction in a shielded area and equipment for handling tritium. The furnaces process irradiated materials such as stainless steel and zircaloy with sample sizes ranging from a single gram to kilograms and containing up to thousands of curies of tritium. The collected gas is analyzed by a high-resolution mass spectrometer.

Automated Process Monitoring and Radiochemical Separations

Staff at the RPL have designed laboratory automation to develop radiochemical process systems to perform a broad range of tasks tailored to both high-activity and trace analyte applications. These include radiochemical analyzers that perform a suite of complex wet chemical functions within closed fluidic systems. We customize these systems to specifically target radionuclides in a wide variety of sample matrices, ranging from high-activity tank waste supernatant to irradiated targets to trace-level analytes in groundwater. Automated instrumentation has been designed to perform in-line sample matrix modification, spike addition, cyclotron target dissolution, column separations, liquid/liquid extraction, and flow-through radiometric or mass spectrometric based quantification.

Applications include automated sample preparation processes for determining trace concentrations of actinides from environmental samples and for designing and developing medical isotope generator systems for yttrium-90 and bismuth-213, as well as isotope purification systems for the cyclotron-produced isotopes zirconium-89 and astatine-211.

Additionally, we have developed radiochemical sensors that can detect and quantify beta-emitting radionuclides in groundwater, such as technetium-99, strontium-90, tritium, X-rays from I-129, and absorbance spectroscopic determination of uranium. Some of the sensors have been integrated into fully autonomous analytical platforms to permit on-site monitoring of well water contamination levels to support subsurface remediation and pump-and-treat processes at contaminated sites.

Spectroscopic On-Line Process Monitoring

From industrial applications to laboratory experiments, on-line monitoring is a valuable tool for real-time measurements of systems enabling immediate feedback for process control and optimization. Analysis output is used in optimizing radiochemical process controls, detecting material diversions, and monitoring fundamental chemical changes. Scientists and engineers at the RPL have developed methodologies for observing and quantifying physical and chemical changes in dynamic systems using commercially available instrumentation. By integrating spectroscopic and physical property measurements with chemometric modeling, RPL staff perform quantitative analyses of a wide array of chemically complex systems.

Spectroscopic on-line monitoring systems, including Raman and UV-Vis/NIR spectroscopy, mass flow/density meters, and temperature probes, are instrumented in banks of centrifugal contactors to perform fundamental and applied research related to nuclear fuel reprocessing. The RPL offers instrumentation for bench-top testing of nonradioactive materials as well as shielded glovebox testing for radioactive materials.

Both systems are available to evaluate a multitude of chemical flowsheets. Other systems for spectroscopic on-line monitoring have also been developed for applications such as Hanford Site tank waste retrieval of both solid and liquid phases and for quantitative analysis of spent nuclear fuel. Efforts include various scales from microfluidic to lab-scale to industrial scale, which spans microscopic for sub nano-liter volumes to telescopic measurements out to 30 meter distances. We apply chemometric models to data collected during operation, which provide real-time information on analyte concentrations and/or process parameters. These methods can be applied to a variety of solid-, liquid-, and/or gas-phase systems.

RPL staff collect real-time spectroscopic data on a flow-through solvent-extraction system for research related to nuclear fuel reprocessing.

Radiological Nuclear Magnetic Resonance Spectroscopy

The RPL's Radiological Nuclear Magnetic Resonance (NMR) Laboratory houses a three-channel spectrometer with a 7.05-tesla wide-bore superconducting magnet and a one-of-a-kind broadband nuclear quadrupole resonance instrument for examining highly radioactive samples, including those containing fissile isotopes. The laboratory provides a broad spectrum of capabilities for analyzing both solid and liquid samples, which in past work have included nuclear waste forms, solution-state uranium complexes, Hanford Site tank wastes, and radioisotope extractant materials. NMR spectroscopy of the technetium-99 isotope has been a uniquely valuable tool in recent projects for the development of Tc separation materials. Other projects use NMR spectroscopy to answer fundamental questions on the structure and bonding of heavy element compounds. In addition, the RPL is the only laboratory in the United States that is currently capable of detecting NMR spectra of the tritium isotope.

The Nuclear Magnetic Resonance Laboratory's instruments offer unique capabilities for research, spanning from fundamental science studies to technology development.

Analytical Chemistry

We specialize in the analysis of highly radioactive materials and very complex sample matrices. Recent materials for research and analysis in the RPL include spent reactor fuel, Hanford Site defense waste, and neutron-irradiated metals and special radionuclides for nuclear medicine. In addition, we routinely analyze water samples, air filters, and smears for radiation monitoring and other project needs. The often complex nature of the materials analyzed calls for customized and adaptable analyses designed by our scientists to meet sponsor objectives.

Nuclear Forensics

Chemical characterization methods applied to nuclear materials and processes for the purpose of attribution form the basis of nuclear forensics–a critically important capability to DOE and national security. RPL tools and techniques were key support for recovery from the Waste Isolation Pilot Plant (WIPP) incident in which a nuclear waste container in storage experienced a thermal excursion event. Commonly used RPL tools for chemical characterization of nuclear materials are optical spectroscopy/microscopy, including IR, Raman and fluorescence spectrometers and microscopes; x-ray diffraction; and fluorescence platforms. Our single-crystal x-ray diffractometer is one of only a few in the nation outfitted with a silver x-ray source ideally suited to penetrate high-Z materials like the actinides. In conjunction with cutting-edge sample containment, we have used this diffractometer platform to discover several new actinide crystal structures. PNNL's capability to rapidly respond during the WIPP incident can support other emergency response scenarios, such as analysis of materials following a nuclear event—e.g., a radiological dispersive device or the Fukushima accident.

Nuclear Materials Examination

Post-Irradiation Characterization

The RPL has capabilities to examine and characterize highly radioactive materials that have undergone irradiation in nuclear reactors. Types of materials examined to date include full-length tritium-producing burnable absorber rods, control rod drive mechanism nozzles from reactor pressure vessel heads, and experimental fuel and targets. RPL researchers and technicians have developed methods for receiving nuclear fuel and waste transport casks, cutting full-length fuel rods for transfer into shielded cells, and preparing irradiated materials for examination. Once the materials are disassembled and subsampled, researchers use a variety of techniques to examine them, including

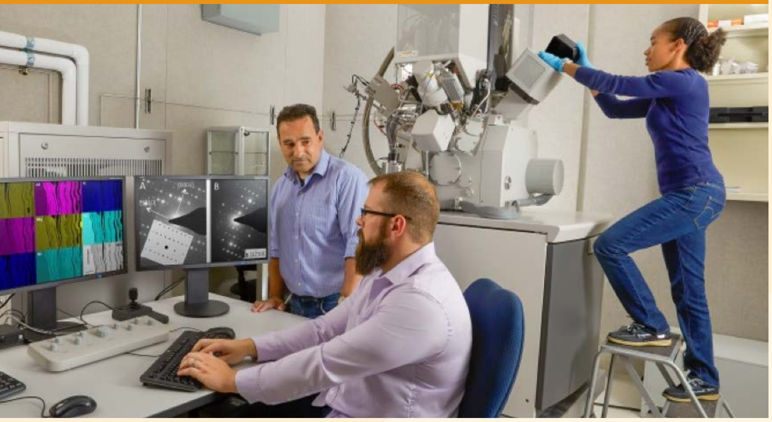
- gamma scanning
- visual examination using high-resolution cameras
- microscopy (optical, SEM, TEM, AFM)
- hydrogen isotope assay (hydrogen, deuterium, tritium)
- ^{3}He assay of steel
- surface analysis (x-ray photoelectron spectroscopy, Auger electron spectroscopy, and secondary ion mass spectrometry)
- thin-film thickness measurements by spectroscopic techniques
- extraction, both gas and liquid
- elemental analysis
- differential scanning calorimetry
- thermogravimetric analysis
- mass spectrometry
- x-ray diffraction.

The RPL staff can perform all of these analyses on classified materials as well.

RPL offers an Instron 8801 servohydraulic multipurpose testing system with a high temperature furnace installed in a modular hot cell that allows

- Testing of irradiated materials at temperatures up to 1100°C in an inert environment
- Tensile, compression, and burst testing of high burnup cladding
- Static fracture, high and low cycle fatigue, and creep-fatigue testing of high temperature reactor materials.

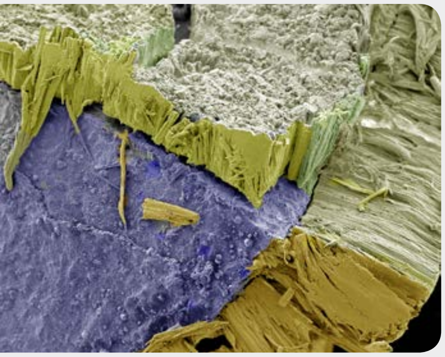
Microanalysis and characterization capabilities, combined with the RPL's ability to receive and prepare highly active and dispersible materials, enable opportunities for radioactive materials research not normally available elsewhere.



The RPL's Helios 660 scanning electron microscope provides unsurpassed insight into actinide chemistry and irradiated metal surfaces and can support projects in heavy-element chemistry, environmental remediation, and radioactive waste management.

Microanalysis and Characterization

At the RPL, we have invested in new electron microscopy technologies for the nuclear sciences, providing unique insights into radiochemistry, nuclear forensics, plutonium processing, and nuclear fuel post-irradiation examination. The RPL Radiological Microscopy Suite provides several new state-of-the-art instruments, including a FEI (Hillsboro, OR) Helios 660 Scanning Electron Microscope (SEM)-Focused Ion Beam (FIB) and a probe-corrected (AC) JEOL (Peabody, MA) JEM ARM300CF Scanning Transmission Electron



SEM image of studtite, a uranyl-peroxide mineral, on uranium dioxide (UO_2). Fundamental studies on the corrosion of UO_2 fuels are important for building the scientific basis for waste disposal.

Microscope (STEM) equipped with the high performance Gatan (Pleasanton, CA) G2™ camera and dual Bruker x-ray energy dispersive (EDS) detectors. These capabilities join a range of other instruments for studying nuclear materials in the suite, including the FEI Quanta 250FEG for working with dispersible actinide and tritiated materials. This microscope is equipped with EDAX EDS, WDS, and EBSD. The RPL also has a range of high-performance optical microscopes (polarized light, metallurgical, and stereo) based in hot cells and contamination areas as well as a full suite of polishing equipment set up in different facilities depending on the radiological hazard.

Spent Nuclear Fuels Challenges

The RPL is uniquely situated to address challenges surrounding the storage and transportation of commercial and defense spent nuclear fuels. Extensive expertise in areas such as fuel oxidation and dissolution, fuel fabrication, advanced processes to dissolve spent fuel and cladding, and materials properties testing, coupled with our irradiated materials characterization and testing capabilities, support development of processing and storage pathways for these highly radioactive materials. Our work with spent and simulated fuels supports U.S. Department of Energy missions ranging from development of geologic repository options for disposal of high-level nuclear waste to extended storage and transportation strategies for these materials, to innovative means for reprocessing and separating key isotopes. The RPL is able to receive commercial spent nuclear fuel rods under the DOE Office of Nuclear Energy's Spent Fuel and Waste Science and Technology Campaign. PNNL performs mechanical tests on these rods to determine the robustness of high burnup spent fuel under conditions of transportation and long-term storage.

Reactor Dosimetry Program

The RPL's reactor dosimetry program measures neutron fluence spectra and calculates radiation damage parameters such as atomic displacements and gas production to characterize complex reactor environments, effectively use material test data, increase reactor safety and plant life extension, develop advanced reactor alloy materials, and design advanced fission and fusion reactors.

In-reactor measurements use custom neutron fluence monitor capsules containing up to 14 materials for the simultaneous measurement of multiple reaction products and for total hydrogen and helium generation. Retrospective reactor dosimetry can obtain similar data using irradiated specimens directly.

The RPL has a comprehensive suite of analytical instrumentation for measuring activation products by gamma spectrometry, alpha or beta counting following radiochemical separations, mass spectrometry on radioactive or stable isotopes, and hydrogen and helium gas measurements.

Nuclear Non-Proliferation Monitoring for the CTBTO

The RPL operates a certified laboratory to analyze air particulate samples for the International Monitoring System (IMS) of the Comprehensive Test Ban Treaty Organization (CTBTO). Samples are received from 80 stations located around the world to look for any evidence of nuclear testing. Our gamma detection sensitivity was recently improved by a factor of 100 with the certification of low-background gamma detectors in the underground lab of PNNL's Ultra Low Background Counting Laboratory and we are also improving the sensitivity of detectors operating in the RPL. The IMS network includes seismic, infrasound, and xenon stations, and PNNL was just certified to operate a xenon gas laboratory in PNNL's Radiation Detection Building. The CTBTO operates 284 monitoring stations worldwide and 183 member states. See ***https://www.ctbto.org/*** for more information.

Shielded Facilities

The RPL contains several fully staffed and equipped shielded cells (or hot cells) for conducting work remotely with highly radioactive materials. The hot cells in the High-Activity Separations Laboratory, High-Level Radiochemistry Facility, Shielded Materials and Examination Laboratory (SMEL), Shielded Process Development Laboratory (SPDL), and Shielded Analytical Laboratory provide unique, complementary capabilities for conducting bench-scale to pilot-scale work with wide varieties and forms of radioactive materials. The four new modular hot cells housed within the SMEL and SPDL are designed to be flexible and configurable to accommodate specific project needs.

Work performed in the hot cells includes analytical chemistry operations, waste tank characterization and process verification, pretreatment, advanced analytical methods development, isotope processing, advanced separations, reactor fuel handling, and nuclear materials examination. Work with classified materials can be conducted in the High-Level Radiochemistry Facility hot cells.

Material examination hot cells housed in the RPL.

Researchers at Pacific Northwest National Laboratory conduct analytical research on nuclear materials using hot cells that shield the researcher from the radiation.

Experience and capabilities inherent in the RPL shielded facilities include

- radiochemical separation and purification
- sectioning of full-length tritium-producing absorber rods for complete post-irradiation examination
- irradiated fuel/target sectioning and processing
- medical isotope production
- thermal processing
- physical properties testing of materials (solid/ liquid separation, centrifugation, settling behavior), including activated metals
- radioanalytical and preparatory chemistry operations (acid dissolution, aqueous/solvent extraction or leaching, distillation, ion exchange, caustic fusion)
- metallography and ceramography
- receipt of spent nuclear fuel rods.

Ensuring Safety, Efficiency and Compliance

The RPL's facility organization works in tandem with researchers to ensure that work is performed safely and efficiently. The RPL is compliant with all applicable regulations pertaining to nuclear safety, quality, environmental safety and health (ES&H), radiological control, and waste management. A state-permitted treatment, storage, and disposal facility is located in the building.

Our integrated operations software is a tool with which to develop, review, and approve research operations authorized under the RPL Routine Operating Envelope. The planning phase for new or revised work includes identifying ES&H hazards and a set of controls tailored to those work hazards. These controls may include permits and approvals, reading assignments, approvals by line managers, and involvement of ES&H subject matter experts.

The RPL also has specific processes that cover all aspects of work performed in a nuclear facility. These processes integrate requirements from the U.S. Department of Energy and other regulatory agencies down to bench-level operations. Our quality assurance (QA) and self-assessment programs work in concert with PNNL operations, ensuring delivery of the highest value to our customers, who dictate the level of QA applied to their work. We have experience working to DOE/RW-0333P and ASME NQA-1 QA requirements.

Special Features of the RPL

- permitted waste treatment, storage, and disposal facility
- low-level waste compactor
- double-shielded, instrumented waste tanks for hot cell use
- remote capabilities to inspect dangerous waste tanks
- continuous monitoring systems to ensure safe operating conditions
- exhaust air sampling capabilities for radioactive material sampling.

U.S. DEPARTMENT OF ENERGY

Pacific Northwest National Laboratory is managed and operated by Battelle for the U.S. Department of Energy.

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About Pacific Northwest National Laboratory

Interdisciplinary teams at Pacific Northwest National Laboratory address many of America's most pressing issues in energy, the environment and national security through advances in basic and applied science. PNNL employs 4,400 staff, has an annual budget approaching $1 billion, and has been managed for the U.S. Department of Energy by Ohio-based Battelle since the laboratory's inception in 1965.

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