Radiochemical Processing Laboratory

Pacific Northwest
NATIONAL LABORATORY

Proudly Operated by **Battelle** *Since 1965*

Housed within the U.S. Department of Energy Hanford Site in south-central Washington State, the Radiochemical Processing Laboratory is a critical facility at the Pacific Northwest National Laboratory, supporting environmental, nuclear, national security, and science missions.

Radiochemical Processing Laboratory

The experienced staff, state-of-the-art facilities, and specialized instrumentation available in the Radiochemical Processing Laboratory (RPL) provide clients with extensive capabilities for process research and development. Capabilities include

- comprehensive nuclear counting instrumentation
- radionuclide separations laboratories
- analytical methods for determining high-concentration to trace-level analytes
- wet chemistry methods
- physical property measurements (such as rheology)
- spectroscopy (UV-Vis, IR, Raman, NMR)
- thermogravimetric and calorimetric analysis
- microscopy (visible light, SEM, TEM, AFM)
- gas and thermal ionization mass spectrometry
- surface science (Auger spectroscopy, EELS, XPS, SIMS).

These capabilities are configured for use with radioactive material in fume-hood, glovebox, and shielded hot-cell environments.

A researcher works with a rheometer to study the fluid-flow properties of complex non-Newtonian media such as solid–liquid suspensions.

Radiochemistry and Processing

Radiochemical Process Development at All Scales

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 researchers work with materials ranging from highly radioactive samples to highly dispersible isotopes to trace levels of radionuclides.

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

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

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

Our glovebox and shielded facility capabilities allow us to design, install, and test small-scale radiochemical processes to support the development of large-scale counterpart facilities, such as the Hanford 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 fluid. Nonradiological waste simulants that possess similar fluid-flow properties have been developed at the RPL to support large-scale mixing and transport tests. A counter-current solvent extraction testing apparatus, equipped with on-line process monitoring capabilities, supports the design and testing of advanced nuclear fuel recycling technologies.

Expert Chemical and Physical Separations

The RPL's unique facilities and multidisciplinary staff enable separations research that extends from the molecular level to the testing of 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 clients' radiological separation and processing needs.

The focus of separations research in the RPL is on testing and experimentation with the actual radioactive materials requiring processing. Capabilities include ion exchange, solvent extraction, and cross-flow ultrafiltration separation

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

techniques. We perform specialized flowsheet testing using 2-cm centrifugal contactors for solvent extraction and a cell-unit filter for radioactive and non-radioactive 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 chemical separation processes.

Separations research in the RPL also encompasses developing new sample processing applications that can 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. Our bench-top to fully contained hot-cell facilities are called into play for thermogravimetric and calorimetric sample analysis.

Plutonium and Tritium Processing

The RPL has capabilities for testing plutonium and tritium processing flowsheets and providing direct design input or solutions for full-scale facilities. Examples include laboratory-scale to bench-top process optimization for the U.S. Department of Energy Tritium Extraction Facility, as well as plutonium conversion and oxidation. Process optimization includes

- determining processing parameters needed to meet facility functional design criteria
- determining input for process flowsheets
- providing input on equipment design or specification
- 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 plutonium and 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.

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.

Plutonium purification and separation processes can be demonstrated in laboratory-scale furnace and dissolution equipment. For the pit disassembly and conversion processes, we demonstrated both gram-size plutonium metal separation from uranium and other metals and reaction with oxygen mixtures. The products from the process are analyzed directly or by methods described in the **Nuclear Materials Examination** section. These capabilities complement the RPL's expert chemical and physical separations capability.

Automated Process Monitoring and Radiochemical Separations

Staff at the RPL have developed automated process monitors and radiochemical separation processes. These include radiochemical analyzers that perform a suite of complex wet chemical functions within closed fluidic systems. Our systems are custom tailored to analyze specifically targeted radionuclides in a wide variety of sample matrices, ranging from high-activity tank waste supernatant to trace-level concentrations in groundwater. Automated instrumentation has been designed to perform in-line sample matrix modification, spike addition, column separations, and flow-through radiometric detection.

Ongoing projects 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, bismuth-213, and zirconium-89.

We have developed automated radiochemical sensors that can detect and quantify beta-emitting radionuclides in groundwater, such as technetium-99, strontium-90, iodine-129, tritium, and uranium. The sensors generally require no reagents to function, so they can be used for in situ measurements of groundwater contamination plumes. These novel systems permit continuous monitoring of groundwater contamination levels to support subsurface remediation and pump-and-treat processes at contaminated sites.

Spectroscopic On-Line Monitoring

From industrial applications to laboratory experiments, on-line monitoring is a valuable tool for real-time measurements of systems when remote observation is necessary or immediate feedback is essential. 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, in which statistics and mathematics are used to extract information from complex chemical data sets, RPL staff perform quantitative analyses of a wide array of systems. Analysis output is used in optimizing radiochemical process controls, detecting material diversions, and monitoring fundamental chemical changes.

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

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 instrument systems are available to evaluate a multitude of 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 used nuclear fuel. 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.

Radiological Nuclear Magnetic Resonance Spectroscopy

Highly radioactive samples, including those containing fissile isotopes, can be examined in the RPL's Radiological Nuclear Magnetic Resonance (NMR) Laboratory. Instruments include a three-channel spectrometer with a 7.1-tesla wide-bore superconducting magnet and a broadband instrument interfaced to a variable-field electromagnet.

The facility is equipped with an array of probes, providing a broad spectrum of capabilities for investigating solid and liquid radioactive samples. NMR research has been performed on nuclear waste forms, solution-state uranium complexes, Hanford Site tank wastes, radioisotope extractant materials, and technetium solids. These solid-state and solution-state experiments are fundamental to structural synthesis and characterization of materials. The RPL is the only laboratory in the United States that can perform NMR measurements of solution-state tritium.

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 the scientists to meet client objectives.

The RPL is the only laboratory in the United States that can perform solution-state tritium NMR measurements.

Radioisotopes for Medical Applications

The RPL excels in ultra-pure radiochemical separations for preparing high-quality radioisotopes that can be used in a variety of medical and scientific applications. We produce generators for lead-212 and bismuth-212 that can be linked to peptides targeting skin cancer (melanoma), breast cancer, and ovarian cancer. We also produce radium-223 that can be targeted to skeletal tumors arising from prostate and breast cancer, as well as thorium-227 that can be linked to monoclonal antibodies for treating leukemia and colon cancer.

Researchers at the Pacific Northwest National Laboratory developed a process to make ultrapure yttrium-90, an important material for radiochemical cancer therapy.

We continue to develop innovative approaches to treating tumors of the liver, prostate, breast, and pancreas (that cannot be surgically removed) using injectable yttrium-90-polymer composites and fast-resorbable seeds. The successful licensing of radionuclide-polymer composite technology to the private sector garnered a Federal Laboratory Consortium Award for excellence in technology transfer. We manufactured the first 20,000 cesium-131 seeds for prostate cancer treatment for industrial partners. Our new zirconium-89 generator will help increase the availability of longer-lived isotopes for high-resolution positron imaging of cancer.

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.

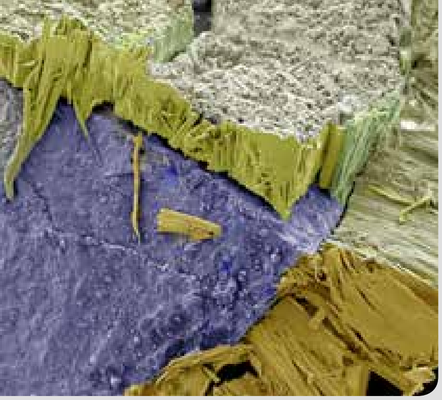
Microanalysis and Characterization

The RPL provides integrated microscopy capabilities related to radioactive materials. The RPL's 300-keV analytical transmission electron microscope provides radionuclide materials analysis at the nanometer scale, almost a thousand times higher resolution than other imaging technologies. This microscope provides researchers with information on a sample's composition, structure, and morphology through spectroscopy, electron diffraction, and imaging.

RPL researchers use the scanning electron microscope with light-element dispersive spectroscopy, wavelength dispersive spectroscopy, and electron back-scattered diffraction capabilities to analyze highly radioactive samples such as spent nuclear fuel. These resources, 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.

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 300-keV analytical transmission electron microscope provides unsurpassed insight into actinide and technetium chemistry and can support projects in heavy-element chemistry, environmental remediation, and radioactive waste management.



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.

Used Nuclear Fuels Challenges

The RPL is uniquely situated to address challenges surrounding the storage and transportation of commercial and defense used nuclear fuels. Extensive expertise in areas such as fuel oxidation and dissolution, fuel fabrication, advanced processes to dissolve used 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 used 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.

Comprehensive Dosimetry Services

The RPL's reactor dosimetry services can help 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.

Our comprehensive services include neutron fluence and spectral measurements, hydrogen and helium gas measurements, and extensive computer calculations of radiation damage effects.

We also provide custom neutron flux monitor capsules containing 14 or more materials for the simultaneous measurement of multiple reaction products and for total helium generation. One small monitor capsule can contain both radiometric and helium gas monitors. Combining multiple monitors into one capsule and providing comprehensive reactor services minimizes the need for reactor irradiation time, providing lower cost and faster service to our clients.

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.

Surface Analysis of Radioactive Materials

The RPL's Radiological Surface Science Laboratory (RSSL) provides a wide range of instruments for examining material surfaces. The RSSL combines these powerful research tools with the ability to examine radiological samples, creating new opportunities for basic and applied research. The RSSL can receive, test, and prepare highly active and dispersible samples for surface analysis.

Surface analysis capabilities include scanning Auger electron spectroscopy, x-ray photoelectron spectroscopy, secondary ion mass spectrometry, and secondary electron and absorbed-current imaging. Depth-profile plots of how element concentrations vary with depth can be collected using each of these techniques.

A researcher prepares to examine surface oxidation of a spent nuclear fuel sample using a SPECS XPS spectrometer mounted on a Physical Electronics 545 Scanning Auger System.

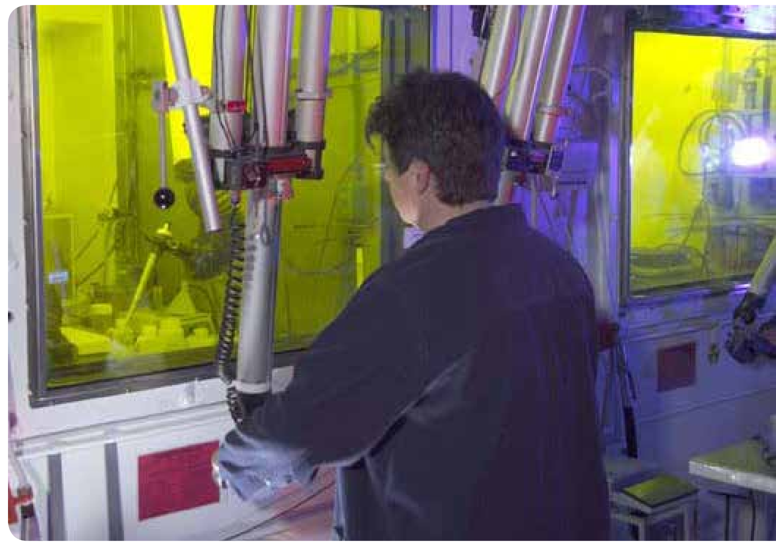
Irradiated Structural Materials Examination and Testing

The Engineering Mechanics and Structural Materials Group operates material examination hot cells within the RPL. The hot cells are used to receive and package radioactive material shipments and prepare specimens for either mechanical property testing or microscopy, which can also be performed in hot-cell environments. For instance, the hot cells host a slow-speed saw for cutting and reducing specimen size as well as polishing equipment for preparing specimens for optical and scanning electron microscopy.

Shielded Facilities

The RPL contains several fully staffed and equipped hot-cell capabilites for conducting work with highly radioactive materials. The hot cells in the High-Activity Separations Laboratory, High-Level Radiochemistry Facility, Materials and Examination Laboratory (MEL), Process Development Laboratory (PDL), 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 MEL and PDL are designed to be 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.



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.

Material examination hot cells housed in the RPL.

Ensuring Safety, Efficiency and Compliance

The RPL's facility oganization 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 onsite.

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 its own 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 federal 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 program alarming and monitoring systems to ensure safe operating conditions
- exhaust air sampling capabilities for radioactive material sampling.

The Radiochemical Science and Engineering Group Manager of the Energy and Environmental Directorate and the Nuclear Operations Division Manager of the Operational Systems Directorate workcollaboratively to deliver world-class research to a diverse customer set.

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 4,500 staff, has an annual budget of nearly $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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