

Tank Waste Processing

Committed to delivering scientific understanding and technological advancements to reduce the risks, uncertainty, time, and cost of resolving critical waste processing challenges—nationally and internationally



The U.S. Department of Energy (DOE) and its contractors rely on Pacific Northwest National Laboratory (PNNL) to provide impactful scientific and technical results to safely complete cleanup of the environmental legacy from nuclear weapons development and government-sponsored nuclear energy research. To meet this need, PNNL is:

- Reducing risks to startup of first-of-a-kind radioactive waste processing facilities
- Identifying and addressing technical gaps and environmental risks
- Enabling waste processing improvements to increase efficiency and reduce mission lifecycle costs
- Providing independent technical bases for near- and long-term environmental cleanup decisions

PNNL directly supports the DOE Office of Environmental Management (EM) mission in four primary technology development and deployment areas:

1. EM Headquarters-led assessments and development of next generation and innovative solutions to reduce cost and risk across the complex
2. DOE Site-office-led research and analysis to understand the challenges and define baseline operational strategies
3. Site-contractor-led technology analysis and maturation to address technical gaps and unanticipated challenges to the baselines
4. Laboratory Directed Research and Development and other PNNL internal investments to understand the fundamental science that technically underpins current applied technologies and the deployment of new innovative solutions

Although DOE-EM is the primary client, PNNL uses its core competencies, state-of-the-art equipment, and advanced facilities to support other national and international clients in search of waste processing technical expertise.





A pool of simulated nuclear waste glass cools after running through the Continuous Laboratory Scale Melter.

STEWARDSHIP CORE COMPETENCIES

To meet the needs of EM's tank waste processing mission, PNNL's research and development portfolio stewards an exceedingly capable and diverse set of staff and capabilities. Over 175 scientists, engineers, and professional staff across the laboratory provide innovative solutions for the nuclear waste challenges of safe storage, retrieval, pre-treatment, immobilization, and disposal. Technical experts, many of whom are internationally recognized leaders in their respective fields, have earned a reputation for excellence, resulting in leadership roles for PNNL on several EM-led partnerships.

These staff underpin the following four core competencies in waste processing: (1) Tank Waste Process Chemistry, (2) Fluid Dynamics and Scaling, (3) Waste Form Development and Testing, and (4) Safety Basis.

PNNL uses its core competencies to solve the problems that are challenging the successful execution of the baseline; resolve high-risk, high-payoff challenges; realize opportunities; and provide the enduring site-specific knowledge to develop and implement solutions to long-term problems.

Tank Waste Process Chemistry

PNNL has performed extensive characterization and process testing on Hanford tank wastes over the last 30 years. This has included receipt and characterization of full core samples under the Tank Waste Remediation System program as well as maturation work in the 1990s and 2000s that underpins the current pretreatment and vitrification flowsheets. Our experience includes maturation of processes for sludge treatment, waste pre-conditioning (dilution, filtration, and evaporative concentration), and cesium and technetium ion exchange (regenerable and single use). PNNL has led these efforts from initial concept through actual waste testing; many unit operations, in conjunction with in-house scaling expertise, are ultimately demonstrated at pilot scale.

Key capabilities include:

- Chemical, physical, and radiochemical characterization and behavior
- Radiochemical and physical separations
- Interfacial and colloidal science
- Chemical kinetics and thermodynamics
- Mineral phase identification
- Actinide chemistry
- Hanford tank waste integrated flowsheet maturation and testing
- Simulant and actual waste testing
- Performance-based simulant development
- Scaled system experimental design

Fluid Dynamics and Scaling

Since the late 1960s, PNNL has demonstrated a robust capability in applied engineering and full-scale, prototypic experimentation in support of process development and commissioning. Our roots reach back to the Hanford Engineering Development Laboratory, and we have supported waste storage, retrieval, feed delivery, and waste treatment on the Hanford Site since the early

production mission. Major contributions in waste processing programs include developing the understanding of mobilization, dissolution, and mixing of salt cake and sludges in waste tanks using rotary jet pumps (waste feed delivery) and pulse jet mixers [Hanford's Waste Treatment and Immobilization Plant (WTP)], jet forces on in-tank components, and slurry transport. PNNL has developed numerical modeling capabilities for turbulent jet mixing of chemically reacting, Newtonian and non-Newtonian slurries. PNNL has supported the technical basis for waste feed delivery to the WTP and treatment process vessels/pipelines using both analytical and numerical models and prototypic or scaled testing.

Key capabilities include:

- Multi-phase (liquid, solid, and gas) flow and transport
- Computational fluid dynamics (advanced modeling for rapid development and application)
- Non-Newtonian yield stress fluids physics and characterization
- Solid/liquid separation and filtration
- Equipment abrasive wear, erosion, corrosion, and erosion/corrosion assessments
- Process assessment via analytical, numerical, and scaled and prototypic testing methods
- Scaling basis and relationships for waste storage and feed delivery unit operations
- Process monitoring technology development, adaptation, and application
- Conceptualization, development, and application of performance-based waste simulants in scaled and prototypic waste processing systems
- Development of simulants for technology and system performance testing
- Double valve isolation performance testing and assessments
- Statistical methods and tools for tank waste sampling and retrieval (rapid development and application)

Waste Form Development and Testing

PNNL has been a leader in waste form development and processing technologies since the late 1960s. Our scientists and engineers developed and applied the joule-heated, slurry-fed ceramic melter to the treatment and immobilization of DOE's radioactive wastes. This work formed the basis for technologies used at the West Valley Demonstration Project, the Savannah River Defense Waste Processing Facility (DWPF), and the Hanford WTP. PNNL's unique application of materials-by-design to nuclear waste glass formulation has enabled real-time design and qualification of nuclear waste glasses within the WTP. We use a science-based approach to develop innovative materials and melter designs to meet specialized, customized waste form needs.

Additionally, PNNL has been a leader in testing and evaluating low-temperature waste forms for the solidification of Hanford tank wastes since the 1980s. PNNL provided process design and waste form development support for the Hanford Transportable Grout Facility and the Hanford Grout Disposal Program. More recently, PNNL has conducted waste form development, testing, and qualification programs for the solidification of WTP aqueous and solid secondary wastes and supporting the evaluation of cementitious waste forms as a supplemental immobilization technology for Hanford's low-activity waste (LAW). Combined with our expertise in tank waste chemistry, PNNL is also developing radionuclide-specific immobilization technologies for key radionuclides such as technetium (^{99}Tc) and iodine (^{129}I).

Key capabilities include:

- Mineral and glass-phase stability and durability
- Simulant and radioactive glass formulation and testing
- Simulant and radioactive low-temperature waste form development and testing



A research team identified a process so a mineral in cement—ettringite—will incorporate and hold the radioactive contaminant pertechnetate.

- Cementitious material and geopolymer formulation development
- Physical and chemical property testing
- Scientific basis for materials performance in disposal environments
- Extensive waste form qualification and performance testing capabilities
- In situ and ex situ irradiation testing
- Composition-property modeling
- Statistical process control and WTP algorithm deployment
- Integration from laboratory-scale to pilot-scale to deployment
- Cold-cap chemistry and melt dynamics testing and modeling
- Scaled-melter systems to assess waste processing rates, cold-cap dynamics, and offgas partitioning including radioactive platform
- Material science and engineering
- Field-scale waste form lysimeter testing in the actual Hanford subsurface
- Expansive modeling capabilities for glass and cementitious waste forms
- Water and reactive transport simulator for near-field performance assessment modeling activities



A sensor the size of a Dixie cup is tracked as it crawls through the tiny air slot underneath the mock-up of a Hanford tank bottom.

Safety Basis

PNNL is a leader in risk assessment, decision science, and associated research and analysis to evaluate and mitigate potential nuclear and chemical safety issues. Our scientists and engineers perform this work for the DOE, U.S. Nuclear Regulatory Commission, commercial nuclear power industry, and international community. This capability is rooted in the Hanford mission, including closure of Hanford tank safety issues, gas generation, retention and release in tank waste slurries, flammable gas, and tank vapors. PNNL holds the Analysis of Record for all double-shell tanks (DSTs) and single-shell tanks (SSTs) at the Hanford Site, consisting of detailed structural analysis of thermal, seismic, and operating load stresses. Our scientists and engineers have developed several ultrasonic testing detection technologies to assess tank leak integrity and inspection applications to improve detection of wall thinning, pitting, and stress corrosion cracking. PNNL is also advancing technologies for deployment in solid phase processing to support the tank life extension program.

Key capabilities include:

- Risk and hazards analysis
- Toxicology
- Tank structural integrity evaluation technologies
- Non-destructive evaluation and analysis
- Ultrasonic instrument design and deployment
- Solid phase processing
- Finite element and structural analysis
- Seismic hazard analysis
- Tank vapor characterization and sensing
- Scaled testing and evaluation of vapor monitoring instrumentation
- Air dispersion modeling
- Flammable gas safety basis assessments
- Gas generation measurements of irradiated samples
- Multi-scale gas retention and release testing

BASIC SCIENCE TO OPERATIONAL SUPPORT

As a DOE Office of Science laboratory, PNNL has delivered and continually develops and enhances its unique capabilities to deliver the scientific and engineering knowledge—from basic science and feasibility research to large-scale demonstrations and plant operations support—that enables maturation and deployment of advanced technologies to achieve EM mission goals. To support technology maturation, PNNL stewards renowned staff, state-of-the-art equipment, and advanced facilities in waste processing that enable integrated, cross-disciplinary understanding and development from the atomic scale through bench and pilot scale as shown below. This suite of capabilities is unique within the national

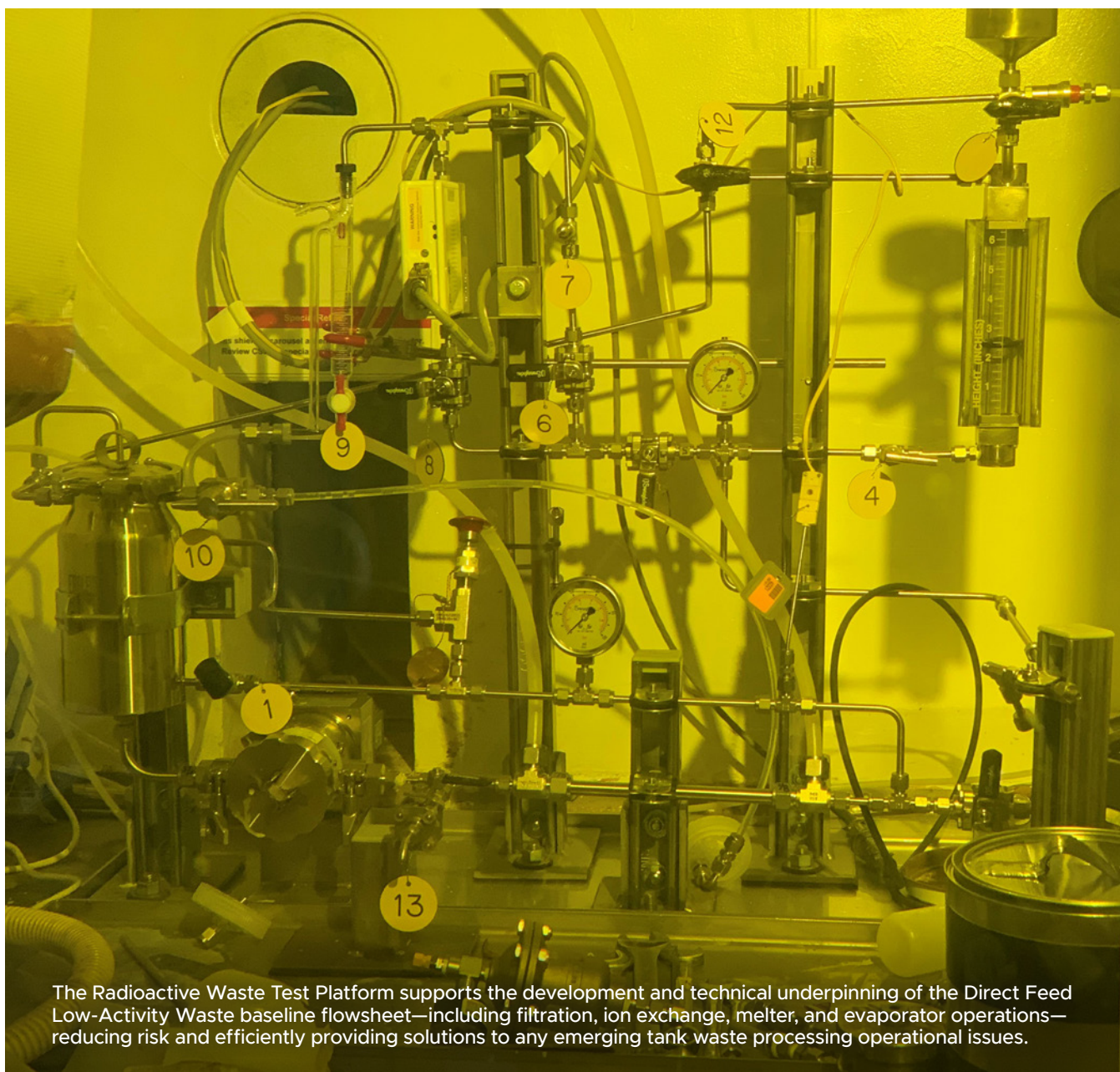
laboratory complex and allows for the continued development of innovative solutions to support deployment and achieve mission goals.

At the heart of our EM mission support is PNNL's Radiochemical Processing Laboratory (RPL), a Hazard Category II nuclear research facility. RPL's mission is to enable research with microgram-to-kilogram quantities of fissionable materials and high-activity materials for developing and deploying innovative radiological material processes and solutions for nuclear waste processing, energy, nonproliferation, homeland security, and isotope production.



One of the RPL's key waste processing assets is the Radioactive Test Platform (RTP). The RTP is a science and technology platform that 1) supports the development of the Direct Feed Low-Activity Waste (DFLAW) baseline flowsheet (the process including filtration, ion exchange, and melter operations), reducing risk for initial and future radioactive operations; and 2) provides a platform to efficiently and effectively address emerging waste processing operational issues.

The RPL's unique facilities, state-of-the-art analytical capabilities, and multidisciplinary staff enable separations research that extends from molecular level conceptualization to testing of prototype flowsheets for industrial applications. PNNL's 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.





KEY OPERATIONAL-BASED ACCOMPLISHMENTS

Tank Waste Storage and Retrieval

- Identified chemical speciation of technetium, iodine, and mercury in tank waste, allowing for development of removal and immobilization technologies and strategies
- Provided technical basis for facility-scale selection for the Hanford Cold Test Facility
- Provided technical basis and support for Defense Nuclear Facilities Safety Board 2010-2 commitments to understand high-level waste (HLW) feed delivery process capabilities
- Provided technical basis and direct operational support for closure of the flammable gas safety issue
- Provided technical basis and direct operational support for remediation and return to service of one of only three DSTs in the 200 West Area
- Provided technical leadership for incorporation of the technical basis for flammable gas retention and release into the documented safety analysis
- Provided technical basis for closure of the retention and release of flammable gas issue in deep sludge layers
- Provided technical basis for safe and effective SST retrievals (e.g., SST sluicing retrievals, SST saltcake dissolution retrievals, retrieval of leaking SSTs)
- Provided technical basis for safe and effective waste feed delivery (e.g., mixer pump configuration and operation, waste transfer configuration and operation)
- Provided tank waste chemistry and toxicology foundation to improve the tank farm industrial hygiene technical basis and transparency of tank vapors information
- Developed conservative correlation for aerosol formation from postulated spray releases



A materials scientist loads single pass flow through test vessels into an oven to test the integrity of the material at various temperatures.

Pretreatment

- Established expected performance of DFLAW filtration system with actual waste and simulant testing to validate proposed design and operational strategy
- Measured actual waste and simulant ion exchange behavior of crystalline silicotitanate (CST) to supply inputs to CST performance models and develop the final Tank Side Cesium Removal (TSCR) column configuration
- Confirmed safety basis assumptions for gas generation to underpin TSCR operations and ion exchange column storage
- Determined that the required TSCR ion exchange column drying time could be reduced by 75%, saving 3 days for each column removal activity on site

- Developed alternative ion exchange conditions to support reduction in nitric acid usage
- Established baseline process for sludge processing, including caustic and oxidative leaching
- Determined mixing requirements for achieving HLW process throughput
- Determined pipeline velocity requirements for sludge pretreatment equipment

Immobilization

- Invented liquid-fed ceramic melter, and transferred technology to other DOE sites
- Lead laboratory supporting design, construction, commissioning, operations, and the deactivation and decommissioning of the West Valley Demonstration Project

- Supported DWPF radioactive operations at the Savannah River Site
- Developed Frit 320 for higher waste loading and melt rate for DWPF
- Developed real-time glass formulation/qualification method for WTP
- Developed baseline glass property models and algorithm for WTP LAW and HLW
- Developed enhanced waste glass property models for DFLAW
- Developed technical basis and scale-up support for the Hanford Grout Treatment Facility, including demonstration disposal vault
- Developed roadmap for the immobilization of Hanford secondary wastes

Disposal

- Established technical basis for Hanford immobilized low-activity waste (ILAW) performance for disposal at the Hanford Integrated Disposal Facility (IDF)
- Provided extensive waste form performance data supporting the IDF performance assessment since the 1990s
- Developed the water and reactive transport simulator to facilitate near-field performance assessments



Radioactive waste from Hanford's AP-105 tank is vitrified into glass in a continuous process during this 2018 demonstration using PNNL's Radioactive Test Platform in the Radiochemistry Processing Laboratory.



One of the many ways PNNL researchers support Hanford Site operations is by running simulated tank waste in a Continuous Laboratory Scale Melter in the Wasteform Development Laboratory—prior to running actual Hanford tank waste through similar equipment in the Radiochemical Processing Laboratory. Pictured on the cover is the viewport where researchers observe the cold cap, which is the slurry that floats on the molten glass before it has fully reacted. *Photo by Andrea Starr | PNNL*

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ABOUT PNNL

Pacific Northwest National Laboratory draws on signature capabilities in chemistry, Earth sciences, and data analytics to advance scientific discovery and create solutions to the nation's toughest challenges in energy resiliency and national security. Founded in 1965, PNNL is operated by Battelle for the U.S. Department of Energy's Office of Science.