Environmental Solutions

A Summary of Contributions for FY04

PNNL-15095

PNNL Contributions to Fluor Hanford

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Fluor Hanford

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National Laboratory
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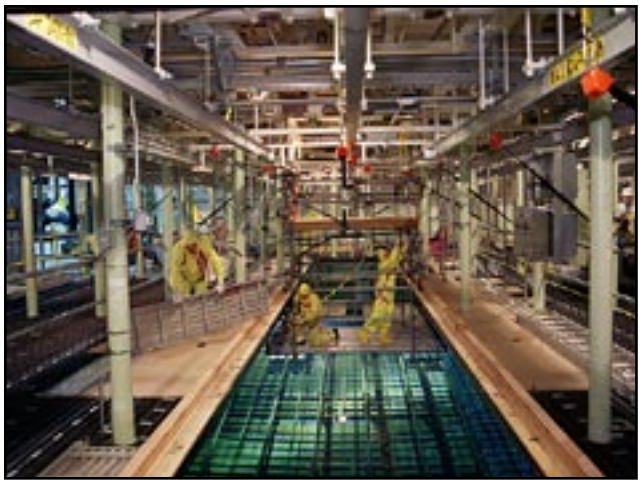
Executive Summary

Partnership Supports Cleaning Up Hanford's Legacy

Pacific Northwest National Laboratory managed a variety of technical and scientific efforts to support Fluor Hanford's work in cleaning up the Hanford Site. Work done for other Hanford contractors, the Waste Treatment Plant, and directly for the U.S. Department of Energy is summarized in the other booklets in this series.



One of the most visible projects at Hanford, the deactivation of the leak-prone K-Basins has been characterized by an unprecedented range of technical challenges.

Removing and Treating K-Basin Sludge

In remediating Hanford's K-Basins, water-filled pools used to store spent nuclear fuel rods, Fluor Hanford is removing years of accumulated radioactive sludge. As part of this effort, PNNL helped develop processes to retrieve and store the sludge. Because the sludge contains uranium that can generate hydrogen gas under certain conditions, PNNL developed a novel approach to predicting the rate at which the gas would be generated. This approach will give Fluor Hanford data to refine its storage options.

With the fuel and sludge removed, Fluor Hanford will pour grout onto the basins' floors to encapsulate contaminants and provide additional shielding. Because some of the equipment in the basins is made from aluminum, which may react with the grout and create hydrogen gas, PNNL studied the chemical interactions for Fluor Hanford.

In the North Loadout Pit of the K-East Basin, Fluor Hanford's plans call for sludge to be retrieved, grouted, and disposed. For this waste, PNNL analyzed several treatment options. After the treatment method was selected, PNNL identified, procured, and tested the equipment necessary to implement it.

Decommissioning Plutonium-Containing Facilities

As more contaminated materials are removed from the Plutonium Finishing Plant and placed in boxes for disposal, Fluor Hanford needs a faster way to determine the radiation level in each box to ensure the proper disposal method is used. To support this requirement, PNNL has developed a quick, non-destructive method to determine the radiation levels for this ongoing effort.

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In response to concerns raised after a fire during a glovebox removal at Rocky Flats, Fluor Hanford and PNNL examined cerium nitrate and other chemicals that would be used in decommissioning PFP. This work was reviewed by the Defense Nuclear Facilities Safety Board, who praised Fluor Hanford for this comprehensive and timely investigation.

In May 2004, Fluor Hanford demolished the 233-S Plutonium Concentration Facility. This was the first DOE plutonium facility to be demolished in open air without being decontaminated to near free-release criteria. At Fluor Hanford's request, PNNL reviewed the atmospheric dispersion modeling, providing data on the location and levels of radioactivity that could be released. Leveraging this experience, PNNL developed estimates of potential releases for demolition alternatives for the 232-Z Waste Incineration Facility.

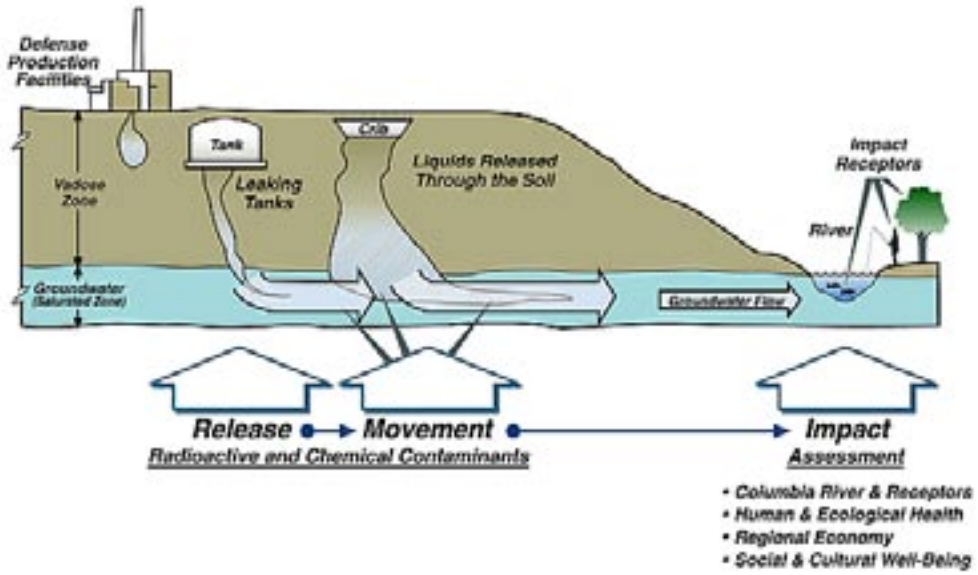
Managing Wastes

PNNL is using air-dispersion models to support planning for future open-air demolition.

As Fluor Hanford has retrieved waste from burial trenches, several large boxes were uncovered. Because of their size, weight, and potential radioactivity levels, the boxes could not be easily moved to the normal radioactive assay system. PNNL adapted the existing techniques and procedures and demonstrated how assays could be completed in or near the trenches. In addition, for Fluor Hanford to maintain its rate of production, PNNL supported developing calibration and operating procedures for the new boxed waste assay systems.

Fluor Hanford is managing a large quantity of plutonium-238, which requires a different disposal method than other wastes. PNNL was part of the team that developed and evaluated options for this problematic waste. In addition to coordinating historical data on the waste, PNNL developed a successful proposal for DOE's Headquarters assistance, needed to bring the plutonium-238's manufacturer into the project.

Fluor Hanford is investigating methods to remove technetium-99 from wastewater, making it easier to treat. As part of this investigation, PNNL tested a series of resins that could be used in ion-exchange columns. In these columns, the waste would pass over the resins, the technetium would be pulled into the resin, and the waste minus the technetium would flow out. PNNL tested one experimental and three commercial resins.



PNNL is supporting Fluor Hanford's work to remediate several sites where the groundwater is contaminated.

Remediating Groundwater

The current method of removing strontium-90 from groundwater in the 100-N Area has proven to be ineffective. At Fluor Hanford's request, PNNL is investigating alternatives for dealing with this contaminant. The first alternative is to use coyote willows to pull the strontium from the groundwater. The second is to form a mineral, apatite, in the aquifer. The strontium is sequestered inside the mineral when it is formed. In addition, PNNL provided and improved access to information systems to support the characterization and continued study of Hanford's groundwater.

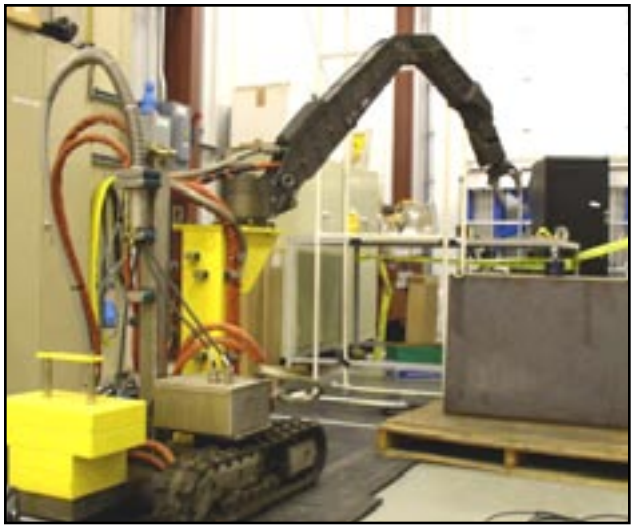
Remediating Soil and Waste Sites

PNNL is supporting Fluor Hanford's work to remediate several sites where the soil is contaminated. For example, PNNL analyzed the impacts of alternatives being considered to accelerate closure of the BC cribs and trenches. Using conceptual and numerical models developed by the Laboratory, PNNL provided Fluor Hanford with information on the fate and transport of certain contaminants.

Contents

Radiochemical Processing Laboratory Inventory Removal – Special Thanks to Fluor Hanford

Thanks to the outstanding contributions of Fluor Hanford, Pacific Northwest National Laboratory (PNNL) was able to remove 63 cubic feet of contaminated equipment and waste material from the Shielded Analytical Laboratory in the Radiochemical Processing Laboratory. Removing this waste reduced the in-cell inventory by an estimated 20 curies of plutonium equivalent and prevented a significant negative impact on hot-cell operations.



The Houdini Track and Predator Manipulator System was used to make a video of the hot cell cleanup operation, greatly reducing the dose and accelerating the cleanout. This system was acquired as surplus equipment from Rocky Flats through a collaborative effort of Fluor Hanford and Battelle.

This work was one part of 17 extremely difficult milestones set for PNNL by the U.S. Department of Energy's (DOE's) Richland Operations Office. The milestones required quick disposal of debris and failed equipment crowding hot cells in the radiochemical laboratory. To meet all 17 of these milestones, all levels of waste management staff at Fluor Hanford and PNNL were involved. They identified waste types and volumes, characterized and designated the wastes, removed the wastes from the hot cells, and shipped the wastes to the Central Waste Complex in the 200 West Area of the Hanford Site. The task was completed on schedule and minimized staff exposure to radiological and industrial hazards.

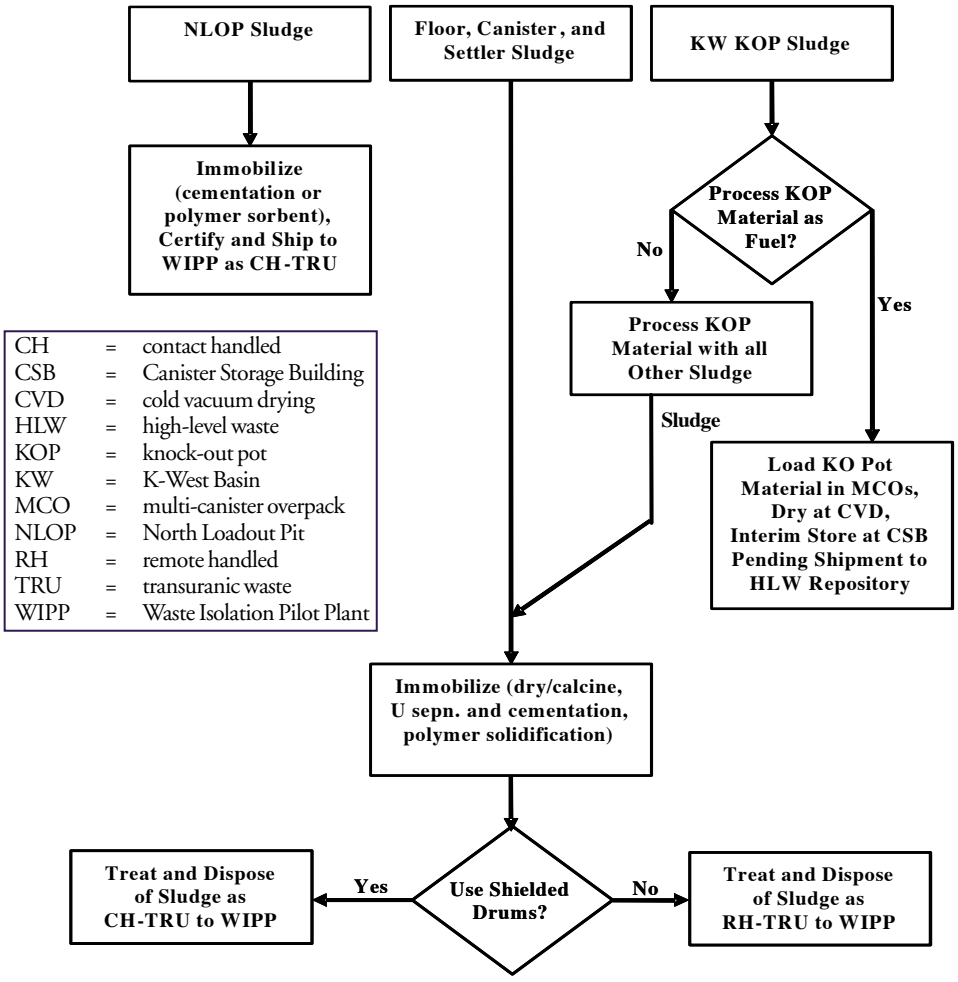
Thanks to the outstanding contributions of Fluor Hanford, PNNL was able to remove 63 cubic feet of contaminated equipment from the Shielded Analytical Laboratory. The task was completed on schedule and minimized worker exposures.

Spent Nuclear Fuel

Options for Dispositioning K-Basin Sludge

To assist Fluor Hanford in removing the spent fuel from the K-Basins and remediating these water-filled basins, PNNL evaluated options for treating and disposing the K-Basin sludge. A key consideration in selecting a treatment process is addressing hydrogen generation from the uranium metal-water reaction ($U + 2H_2O \rightarrow UO_2 + 2H_2$). Two groups of treatment options were considered – those that would be performed at ambient temperature (such as grouting) and those that would require elevated temperatures (such as calcination or bulk vitrification). The higher-temperature options would oxidize the uranium metal and thereby prevent hydrogen generation.

Options were evaluated for treating and disposing of K-Basin sludge.



A disposition strategy for the K-Basin sludge, based on the evaluation of sludge disposition options presented in PNNL-14729, shows separate disposition pathways for North Loadout Pit sludge; floor, canister, and settler sludge; and knock-out pot sludge.

Key conclusions of this study included the following:

- Ambient temperature processing would be preferable to higher-temperature processing due to reduced process complexity.
- Because the sludge in the North Loadout Pit (NLOP) in K-East Basin contains a much lower uranium metal concentration than the remainder of the sludge, uranium metal removal would not be required before use of ambient temperature processing such as grouting for this sludge fraction.
- However, for the majority of the K-Basin sludge, use of ambient-temperature treatment would require demonstrating one of two conditions
  - The solidification matrix (for example, grout) essentially eliminates the reaction between the uranium metal and water that will be present in the treated sludge, or
  - The rate of the uranium metal-water reaction in the solidified sludge is substantially lower than the rate of reaction in oxygen-free water, **and** removing the majority of the uranium metal present in the sludge as a preconditioning step before solidification.

The results of PNNL's study for Fluor Hanford are documented in *Disposition Options for Hanford Site K-Basin Spent Nuclear Fuel Sludge* (PNNL-14729), issued in January 2004.

K-East Basin North Loadout Pit Sludge Treatment

In fiscal year (FY) 2004, PNNL supported Fluor Hanford's effort to develop the treatment process and disposition path of the sludge in the K-East Basin NLOP. Plans call for this sludge to be treated by grouting and then packaged for disposal at the Waste Isolation Pilot Plant as contact-handled transuranic waste. PNNL characterized samples of NLOP sludge and evaluated various waste form and package configurations for the treated sludge. This work was documented in *Evaluation and Recommendation of Waste Form and Packaging for Disposition of the K-East Basin North Loadout Pit Sludge* (PNNL-14741) issued in January 2004. Grouting in 55-gallon drums was selected as the treatment method. PNNL identified, procured, and tested the equipment required to implement this treatment method, developed drafts of the required operating procedures, and completed other activities required for installation and operation of this equipment. PNNL conveyed to Fluor Hanford documentation showing the following: 1) the design of the PNNL sludge treatment system, 2) the testing that had been performed, and 3) the functional requirements that were met. The equipment was transferred to Fluor Hanford in early FY05.

Sludge in the North Loadout Pit of the K-East Basin was characterized, and various waste-form configurations were evaluated for the treated sludge.

Fluor Hanford plans to install the grout system in T-Plant. PNNL will continue to support Fluor Hanford during installation at T-Plant, system startup, and sludge treatment.

As a result of the Fluor Hanford /PNNL collaboration, Fluor Hanford will immobilize the 6.3 cubic meters of NLOP sludge some 30 years earlier than originally scheduled. This immobilization effort could be an important demonstration of the treatment for other sludges.

The process of grouting sludge from the North Loadout Pit in 55-gallon drums was tested.

The grouted waste was removed from the drum and tested.

Treatment of the Remaining K-Basin Sludge

Testing showed that grouting alone is not sufficient to treat the K-Basin sludge.

Because of significant uranium metal concentrations in the remainder of the sludge in the K-Basins, the impacts of uranium metal need to be considered selecting the treatment process or processes. To support Fluor Hanford in final treatment of the K-Basin sludge, PNNL conducted gas generation testing of uranium metal in K-Basin sludge and in grouted waste forms. The testing showed that grouting alone is not sufficient to treat the K-Basin sludge. Additional or alternative treatments will be required to substantially reduce the hydrogen gas generation rate. These treatments might include reacting (oxidizing) the metallic uranium, possibly with a preliminary uranium metal separation/concentration step; removing the water from the sludge by decanting and drying; or solidifying the sludge with Waste Isolation Pilot Plant-acceptable agents to coat the reactive uranium metal surfaces. The PNNL document *Final Report – Gas Generation Testing of Uranium Metal in Simulated K-Basins Sludge and in Grouted Sludge Waste Forms* (PNNL-14811) was issued on August 18, 2004.

PNNL conducted gas-generation tests of uranium metal in grouted K-Basin sludge. Several Portland cement forms are shown here.

To determine the technical feasibility of using commercial gravity concentration equipment for segregating uranium metal in K-Basin sludge, PNNL coordinated and summarized testing performed by two equipment vendors. The vendors used a sludge simulant that contained cobalt-cemented tungsten carbide as a uranium metal surrogate. In this testing, more than 96% of the uranium metal surrogate was concentrated into 10% to 30% of the sludge mass (7% to 24% of the sludge volume). With more prototypical equipment and stream recycle, higher recoveries may be achieved.

Commercial equipment was tested for segregating gas-generating uranium metal in K-Basin sludge.

For K-East Basin floor, pit, and canister sludge, the test results indicated that gravity segregation, in conjunction with solidification in a tailored grout matrix, can result in a waste form for disposal at Waste Isolation Pilot Plant, for which waste loading will not be limited by the rate of hydrogen generation from the uranium metal-water reaction.

For K-West Basin canister sludge, gravity segregation will yield a uranium-depleted stream for which the remaining uranium metal will be slightly more constraining than the plutonium-239 fissile gram equivalent constraint for Waste Isolation Pilot Plant disposal.

Gravity segregation processing of the knock-out pot sludge is not recommended. Without uranium metal removal, the knock-out pot sludge is expected to exhibit a uranium metal content in excess of 20 wt%. It is unlikely that use of the equipment evaluated in this study could achieve the 99+% uranium removal that would be required for this sludge fraction.

The results of the demonstrations are summarized in PNNL document *Segregation of Uranium Metal from K-Basin Sludge: Results from Vendor Testing* (PNNL-14845), issued in September 2004.

The Gekko Systems InLine Pressure Jig, Model IPJ600, (left) and Knelson Laboratory-scale Batch Concentrator, KC-MD3, (right) were tested to determine whether or not they could be used to segregate uranium metal in K-Basin sludge.

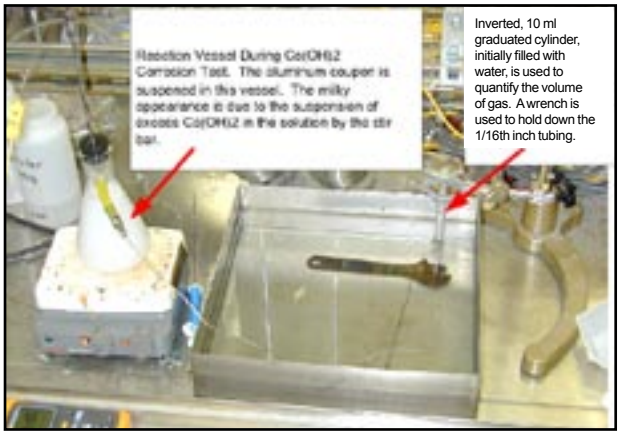
K-Basin Deactivation and Decommissioning

Fluor Hanford's current end point for the K-Basins will require grouting the basin floors after the sludge and fuel have been removed to encapsulate contaminants and equipment. The grout will also provide shielding from dose. Some of the equipment items in the basin are made from aluminum, which is expected to react with the high-pH grout, generating hydrogen. At Fluor Hanford's request, PNNL performed two studies to support addressing the grout hydrogen-generation problem.

Aluminum Debris in Grouted Basins

The potential impact of encapsulating aluminum equipment items with grout in the K-Basins was assessed.

PNNL assessed the potential impact of encapsulating aluminum equipment items (canisters, identification tags, poles, etc.) within the planned grout pour in the K-Basins. A letter report was prepared based on an experimental and calculational evaluation of the aluminum/grout reaction. The report concluded that 1/8th of the estimated aluminum inventory in the K-East Basin would have to be removed to ensure the hydrogen concentration remained below 25% of the lower flammability limit (1% hydrogen).



Using calcium hydroxide and aluminum in this test setup, PNNL researchers measured the amount of hydrogen that would be generated by encapsulating aluminum equipment items within the planned grout pour in the K-Basins.

Aluminum Analyzer

Given that aluminum items will need to be removed from the K-Basins and that visual confirmation of metal types may be difficult due to corrosion, a non-destructive evaluation method to discriminate between aluminum and stainless steel is needed. PNNL developed and deployed an underwater non-destructive sensor using a technology originally designed to enable border inspectors to differentiate between metals of similar appearance. The canister segregation instrument employs eddy current technologies to make on-contact electrical conductivity measurements in 2 to 3 seconds.

K-East Basin Sludge Flocculant Testing

Seven commercial flocculants were tested to identify those with the best performance basis on K-Basin sludge.

To isolate the sludge from the basin, Fluor Hanford will transfer the K-East Basin sludge from the floor and pits to large, freestanding containers, located in the pits. When the sludge is pumped into the containers, it must settle and clarify quickly to prevent radioactive material from being suspended in the overflow water returned to the basin, which could cloud the basin water and increase the dose rate to the operations staff. PNNL scientists have been working to enhance sludge settling and clarification by adding a flocculant to the sludge while it is being transferred to the containers.

PNNL tested seven commercial flocculants with a simulated K-East Basin sludge to identify those with the best performance characteristics. Actual sludge was then used for flocculation testing in a hot cell. From this testing, PNNL researchers determined that a cationic polymer flocculant called Nalco Optimer 7194 Plus exhibited superior performance.

Nuclear Safety Support to K-Basin Decontamination and Decommissioning

PNNL is providing nuclear safety and licensing support to Fluor Hanford for activities at the K-Basins, as required by *Code of Federal Regulations*, Title 10, Part 830, "Nuclear Safety Management." In FY04, PNNL developed safety basis cases for sludge recovery and containerization, transfer of sludge from the K-East Basin to the K-West Basin, grouting K-Basin pits, and decontamination and decommissioning (D&D) of the K-East Basin. In addition, PNNL determined the initial potential public and worker risk from new activities at the K-Basins and identified a risk management approach to meet DOE risk management criteria. To do this, PNNL

Safety basis cases were developed for K-Basin radioactive sludge recovery, containerization, and transfer.

- Performed hazard categorization, hazard analysis, and accident analysis
- Identified and assessed the effectiveness of nuclear safety related controls (both hardware and procedural)
- Produced safety basis and supporting technical documents, per DOE and Fluor Hanford requirements.

PNNL also provided technical support for the development of safety-related technical positions, K-Basin safety basis amendments, Unreviewed Safety Question evaluations, and linkages with DOE's Richland Operations Office licensing staff and the Defense Nuclear Facilities Safety Board. This work is just part of PNNL's ongoing efforts to support K-Basin nuclear safety.

As part of its ongoing efforts to support K-Basin nuclear safety, PNNL developed safety basis cases for sludge recovery and containerization.

Method for Predicting Gas Generation by Uranium-Metal-Bearing Hanford K-East Basin Sludge

In FY04, PNNL developed a novel approach to predicting gas generation by uranium-metal-bearing sludge with specific application to Hanford K-East Basin sludge. This method accounts for production of hydrogen gas and oxidizing species by radiolysis, hydrogen production by chemical reactions, and consumption of oxidizing species by chemical reactions, with appropriate transitions and dependencies on temperature, dose rate, and concentrations of species. The method is applicable to other waste forms provided that principal reactions can be determined.

Uranium-metal-bearing sludge from the K-East Basin is to be loaded into large-diameter containers (LDCs) and moved to interim storage in a dry cell at T-Plant. Chemical behavior of sludge in LDCs is of interest to design and safety because both radiolysis and oxidation of uranium metal generate hydrogen gas, with resulting implications for flammability of the container and cell headspaces.

The new model explains observed laboratory behavior, including so-called incubation times for the uranium metal-water reaction, and can be readily applied to proposed storage processes. Its key feature is to separately define the production rates of specific gases by radiolysis in interstitial water and consumption rates of oxidizing species by solids. Uncertainty in experimental data is derived and applied to each rate law, providing a basis for controlling an efficient, effective process to react the metallic component of metal-bearing sludge to form more stable uranium oxide sludge with reduced hydrogen generation rates. This more stable sludge is more amenable to grouting, disposal, and storage. The cost of disposal and risk of storage are significantly reduced over the cost and risk for metal-bearing sludge.

A novel approach to predicting gas generation by sludge containing uranium metal was developed.

Assessment of Viability, Schedule, and Cost for Processing Sodium-Bonded Fuel Pins from the Fast Flux Test Facility

As a part of activities to close and dismantle the Fast Flux Test Facility, Fluor Hanford was faced with the disposal of sodium- and NaK-bonded fuel pins. This unique fuel poses special handling and disposal challenges, because of the high chemical reactivity of the elemental sodium. Fluor Hanford requested that PNNL assess the viability of processing the Fast Flux Test Facility's sodium and NaK-bonded fuel pins using the Radiochemical Processing Laboratory. Several processing options were evaluated, and direct oxidation was selected as the preferred alternative based on process simplicity and acceptability of the product by the repository. Following the study, Fluor Hanford learned that direct disposal of the fuel was under consideration by DOE Headquarters, which would effectively obviate the need for processing.

Plutonium Finishing Plant (PFP)

Glovebox Decontamination and Secondary Wastes

To support Fluor Hanford in decontaminating the plutonium-containing gloveboxes at PFP, PNNL and PFP investigated the chemical reactivity hazards of the secondary wastes generated by three candidate decontamination technologies: cerium nitrate/nitric acid, EAI Corporation's RadPro®, and AREVA Corporation's Glygel.

PNNL and PFP investigated the chemical reactivity of the secondary wastes created by three possible technologies to decontaminate plutonium-containing gloveboxes.

When dried, the cotton rags used to wipe up spills of cerium nitrate decontamination solution showed a low-rate exothermic reaction at 30-40°C that could lead to waste drum overheating. This heating results from reactions between nitric/nitrate ions in the cerium nitrate solution and cellulose in the rags. Further evaluation showed that the addition of moisture to the rags will absorb the energy of the exothermic reactions and prevent the waste drums from overheating. Additional investigations showed that using commercially available synthetic (polyester-nylon) rags prevented the low-temperature exothermic reactions from occurring.

The thermal reactivity of this neutralized and reduced ceric nitrate/nitric acid soaked cotton rag was tested.

Testing of the RadPro® decontamination solutions demonstrated their safety and also showed that neutralizing the solutions dramatically increases the safety margin. Future testing is planned for AREVA Corporation's Glygel product. A PNNL technical report will be prepared in FY05 to summarize the results of all the testing and present the recommendations.

Chemical Reaction of Leaded Rubber Gloves with Nitric Acid

Some of the gloveboxes in PFP have not been used for many years, and questions were raised by project managers regarding the safety of leaded gloves that may have previously been in contact with nitric acid. Two documented incidents have occurred: 1) an explosion at the Mound facility during heating of gloves in a muffle furnace when flammable vapors ignited, and 2) a minor fire in a drum of leaded gloves at Rocky Flats. For Fluor Hanford, PNNL reviewed and summarized the results of a number of studies regarding the chemical reactions with the gloves.

Based on studies of the chemical reactions of PFP gloves with nitric acid, aging gloves will be replaced as the first step in decommissioning certain PFP gloveboxes.

Leaded gloves are produced by sandwiching red lead oxide between two layers of rubber, which can be a laminate of hypalon (a synthetic rubber) and neoprene (a polymer of chloroprene). If such a laminate is used, a bonding agent is needed to adhere the hypalon to the neoprene. When leaded rubber gloves are in contact with nitric acid, degradation of the rubber and the lead occurs.

PNNL's analysis showed leaded rubber gloves exposed to nitric acid form degradation products that can exhibit explosive behavior at temperatures exceeding 170°C. These degradation products are also shock sensitive, with relatively low impacts resulting in explosive behavior. A simple aqueous wash of the degradation products stabilizes the product against these explosive reactions. Neoprene must be present in leaded rubber gloves for these reactions to occur. Hypalon does not degrade in nitric acid and does not contribute to these explosive reactions. As a result of this study, project managers decided to replace aging gloves as a first step in the decommissioning work.

Robotics Support to Plutonium Reclamation Facility

To support Fluor Hanford's decommissioning of PFP, PNNL staff organized a brainstorming and planning session to discuss methods and approaches to clean the floor in the Plutonium Reclamation Facility canyon. Nationally recognized experts in robotics and decommissioning were invited from other DOE sites, offering proven approaches and insights to this challenge. PNNL staff presented information on equipment that was available at the time for use on the project as well as Laboratory capabilities.

PNNL staff participated in a brainstorming session to find options for cleaning the contaminated floor of the Plutonium Reclamation Facility.

Duct Cleaning and Fixative Application

As part of Fluor Hanford's work to remove a large amount of legacy plutonium holdup in a 10-inch-diameter ventilation duct at PFP, PNNL identified potential concepts for the remote removal equipment. In addition, PNNL built a mockup of the ductwork at the Volpentest HAMMER Training and Education Center. This resource is expected to be used to develop and demonstrate tools and methods needed to remove plutonium from ductwork, and to prepare ducts for demolition activities. In addition, the system will likely offer an appropriate environment for operator training.

PNNL is using this mockup to evaluate ideas to remove radioactive contamination, or holdup, in a 10-inch-diameter section of ductwork at PFP.

Qualification of Assay Systems for Standard Waste Boxes at PFP

As Fluor Hanford gears up to decommission PFP, they will begin generating copious quantities of contaminated waste. Historically, wastes from PFP have been packaged in 55-gallon drums, but that practice requires significant effort to cut equipment into pieces small enough to fit. A more efficient approach adopted in Fluor's plans will rely on using standard waste boxes for most of the waste. However, there was no procedure to assay these large boxes to meet the requirements of PFP Safeguards.

PNNL was tasked by Fluor Hanford to develop a standard procedure to assay large standard waste boxes at PFP. The contents of the standard waste boxes could include a range of materials, including contaminated paper and rags, valves, piping, and equipment. Determining the quantity of radioactive contamination in these boxes presents many technical challenges, considering the gamma and neutron attenuation caused by the contents of the boxes. PNNL's approach utilized a neutron slab counter for transuranic material, and an ISOCS gamma spectrometer to detect and identify gamma-emitting radionuclides.

PNNL used a neutron slab detector to determine the radioactive contamination in standard waste boxes filled with materials from the cleanout of PFP gloveboxes.

A new technique was developed to assay large standard waste boxes at PFP. The boxes will be used in removing materials from the aging plant.

With the appropriate counting strategies, configuration, and software, the new technique met PFP's rigorous requirements for assaying boxes.

To demonstrate the effectiveness of assay systems for standard waste boxes, measurements were performed on a box mockup using radioactive sources placed in the seven pipes, which were surrounded with a variety of simulated waste materials.

PNNL demonstrated the effectiveness of the assay systems by constructing a mockup of a standard waste box fitted with an insert that included sample pipes in seven different positions. These sample pipes were selectively loaded with standard radioactive sources. The spaces between the sample pipes were filled with rags and surplus metal equipment to simulate the loading of a PFP standard waste box. Measurements were performed with a variety of sources in the seven pipes, and a variety of filler materials in the box. The results of these measurements helped PNNL select appropriate counting strategies, including detector positions and counting times, and helped them qualify their analysis software. With appropriate counting strategies, configuration, and software, PNNL researchers were able to demonstrate that their assaying technique could meet the rigorous requirements for assaying the boxes set by PFP Safeguards. Qualifying the standard waste box counter was a critical step in preparing for the cleanout of PFP gloveboxes.

Deactivation and Decommissioning (D&D) Activities

PFP D&D Team Efficiency Review

To remove legacy materials and gloveboxes from PFP in FY05, Fluor Hanford will be dramatically increasing the number of staff involved, from 5 to 30. Before this happens, Fluor Hanford wanted an independent review of the D&D work practices to determine if there might be opportunities to improve efficiency. To support this effort, a PNNL team consisting of an industrial engineer and an engineer with extensive international nuclear D&D experience observed work practices at PFP during the fall of 2004.

PNNL conducted an independent review of the D&D work practices at PFP to determine if there might be opportunities to improve efficiency in FY05.

An independent review of the D&D work practices at PFP was conducted to determine if there might be opportunities to improve efficiency.

The PNNL team reviewed documentation of Fluor Hanford work practices. They also observed workers during pre-job briefings, doing work in contaminated spaces, post-job reviews, and an ALARA committee review. Finally, they interviewed workers individually and in group settings. Recommendations from the analysis included changes to work planning activities as well as the makeup of work team skill sets.

Technical Support to Airborne Modeling for 233-S Facility Demolition

In FY04, Fluor Hanford demolished the 233-S Facility, which was used to concentrate plutonium nitrate. This was the first open-air demolition of a highly contaminated plutonium facility at the Hanford Site, and within the DOE complex. For this reason, very little empirical information existed on the quantity of radioactive material that was likely to be released from demolition activities. Information on the effectiveness of release mitigation techniques was equally scarce. At Fluor Hanford's request, PNNL assisted with pre-demolition planning and review of atmospheric dispersion modeling, which was used

to provide information on the location and levels of radioactivity that potentially could be released. In addition, PNNL provided contamination dispersal estimates. This information helped support the first-ever open-air demolition of a highly contaminated facility.

In May 2004, Fluor Hanford completed Hanford's first-ever open-air demolition of a highly contaminated facility with assistance from PNNL.

Airborne Contamination Modeling for 232-Z Facility

By 2006, Fluor Hanford plans to demolish the 232-Z Facility, part of Hanford's plutonium recovery efforts. This is the second demolition of a contaminated plutonium facility planned at the Hanford Site. Leveraging the experience from the 233-S Building demolition, PNNL developed engineering estimates of potential releases for demolition alternatives. In addition, PNNL conducted atmospheric dispersion modeling using those release rates to provide information on the location and levels of radioactivity.

In this modeling, PNNL used a computer code developed by the U.S. Environmental Protection Agency that calculates dispersion patterns considering building wake effects and other meteorological phenomena. This work is influencing the amount of residual contamination that may be allowed in the building as well as the overall approach and demolition techniques. During this effort, PNNL and Fluor Hanford have interacted frequently as interim results are generated. Fluor Hanford will use the results to plan demolition activities that will keep potential contamination within the limits established for the project contamination area.

Waste Stabilization and Disposition

Technical Consultation on Plutonium-238 Disposition

Options were developed and evaluated to dispose of the plutonium-238 on the Hanford Site.

A significant quantity of plutonium-238 was sent to Hanford in the late 1960s for research to determine the criticality properties of that isotope. However, the additional plutonium-238 needed to perform the experiments was never made available, so it was placed into long-term storage in the original shipping containers. This plutonium isotope is much more hazardous to handle than most of the plutonium produced at Hanford because it produces a very high neutron flux. In addition, the high concentrations of plutonium in this material required a disposition path different from other retrievably stored transuranic waste. To support developing this pathway, PNNL provided a report with historical information on the material and worked with the Savannah River Site, the manufacturer of the material, to obtain the original shipping records and other process characterization data. To support development of a more detailed disposition plan, PNNL and Fluor Hanford developed a technical assistance proposal that was approved by DOE's Headquarters. The Fluor Hanford/PNNL team selected a preferred disposition path for the material from several alternatives that were identified by the team.

Cost and Risk Evaluation of a Remote-Operated Size-Reduction System for T-Plant

At DOE's request, the Remote-Operated Size-Reduction System was evaluated for possible deployment in the T-Plant Complex. Based on the minimal amount of risk avoidance and the significant costs, the system was not recommended.

Fluor Hanford was asked to consider the benefits of using the Remote-Operated Size-Reduction System, a self-contained system to process transuranic-contaminated gloveboxes and tanks and package them into standard waste boxes. The system was designed for use at the Rocky Flats site, but after testing, it was never used. Fluor Hanford assembled a team including PNNL to evaluate the Remote-Operated Size-Reduction System for possible deployment at the T-Plant Complex.

The team was initially asked to consider the following deployment options:

- Stand-alone outdoor deployment with integral high-efficiency particulate air (HEPA) filters
- Stand-alone outdoor deployment with exhaust tied to the 2706-T stack
- Assembled in 2706-T or TA bays with exhaust tied to the 2706-T stack
- Assembled in 221-T head-end with exhaust tied to the canyon ventilation system
- Assembled on the canyon deck with exhaust tied to the canyon ventilation system.

The stand-alone outdoor options were eliminated because the system is not designed to operate outdoors. The 2706-TA bay and the 221-T head-end options were eliminated because the system dimensions are too large for it to fit without extensive modifications. The team evaluated use of the unit only in the 2706-T bay and on the 221-T canyon deck.

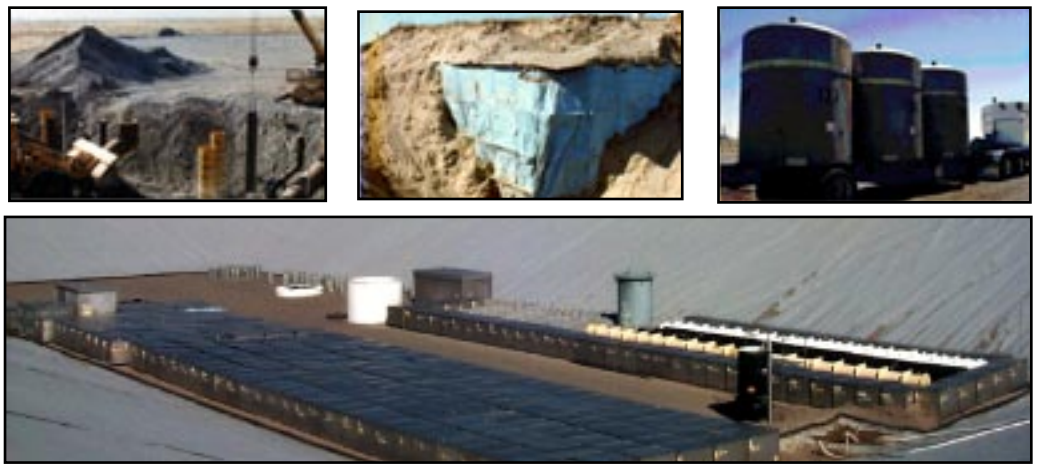
The team recommended that management take no action to acquire the ROSRS for deployment at T-Plant based on a minimal amount of risk avoidance that would be gained at a significant cost.

Planning Basis for M-91 Waste Processing

A new, phased approach was recommended to process the M-91 wastes. The new approach would complete the waste processing nearly a decade ahead of schedule.

Fluor Hanford is working to accelerate processing of Hanford Site transuranic waste and mixed low-level waste that cannot be processed using existing Site capabilities. These wastes include both oversized and remote-handled mixed low-level waste and transuranic containers, that is, containers with surface contact dose rates greater than 200 mrem/hr. These wastes are called M-91 wastes because their processing is required in *Hanford Federal Facility Agreement and Consent Order* (TPA) milestone M-91-01.

In supporting Fluor Hanford in this effort, PNNL has recommended a new, phased approach that would complete the M-91 waste processing mission nearly a decade ahead of the TPA-mandated schedule. In addition, PNNL's accelerated processing of M-91 wastes is predicted to save nearly $125 million in life-cycle costs by leading to the early retirement of T-Plant.



PNNL has recommended a new, phased approach to support completing the M-91 waste processing mission nearly 10 years ahead of the Tri-Party Agreement's schedule.

Analysis and Recommendations for Gamma Guard® as a Waste Additive

At Fluor Hanford's request, PNNL confirmed the results of an earlier analysis by Duratek and supported their conclusion that Gamma Guard®, a non-leachable leaded-glass product, would reduce dose rate to workers or, conversely, that it would allow additional activity to be added to a waste container while maintaining a constant shielding effectiveness. In addition, PNNL noted that the additive looks promising for shielding applications for Hanford wastes containing gamma emitters (e.g., cesium-137). Thus, applications like K-Basin sludge retrieval, transport, and processing might benefit from using Gamma Guard®.

In addition, PNNL identified another potential K-Basin application. Ion exchange modules are currently used at the K-Basins to remove cesium from the water, and these modules also pick up alpha materials. Care must be taken to avoid picking up too much alpha radiation and making the modules transuranic waste. However, if Gamma Guard® was added to the grouted waste in the drums, the greater mass would allow the modules to contain more alpha material without exceeding the transuranic threshold. This would translate to less frequent replacement of the ion exchange modules, which should reduce costs and save time.

At the conclusion of their study, PNNL recommended an engineering study to more accurately determine the benefits of Gamma Guard® for the identified applications.

Support Deployment of New Standard Waste Box Assay Systems

To assist Fluor Hanford in assaying standard waste boxes, PNNL is developing an operating procedure for the Super High-Efficiency Neutron Coincidence (SuperHENC) Counting System, manufactured by BNFL Instruments, Inc. This operating procedure will be applicable for the entire Hanford Transuranic Waste Program at Waste Receiving and Processing Facility, PFP, and elsewhere on the Site, as needed.

The SuperHENC is a self-contained mobile transuranic waste assay system mounted on a commercial trailer designed for rapid deployment. The first third of the trailer houses the control room, the mid-section houses the passive neutron coincidence counting system, and the final third houses a Gamma Energy Analysis System for the quantification of required radionuclides. Waste packages are loaded onto a fold-down drawbridge with an integrated load cell and a motorized pallet to introduce the sample into the assay chamber, which will hold drums or a standard waste box up to 37 inches high by 65 inches wide by 71 inches long.

A procedure to operate the self-contained mobile transuranic assay system is being developed. The procedure will apply to the entire Hanford Transuranic Waste Program.

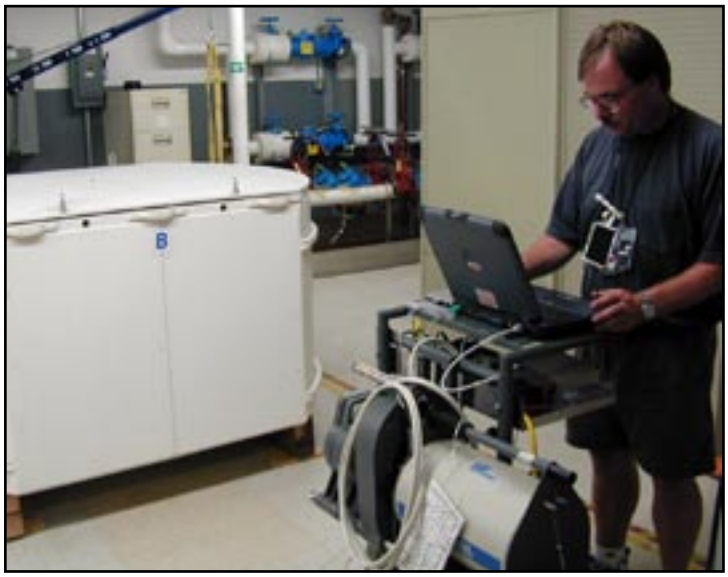
The system is optimized for the sparse counting signal characteristic of large packages and dense matrices. The software automatically integrates the SuperHENC neutron data with the gamma data collected, and calculates the sample-specific lower limit of detection. All Waste Isolation Pilot Plant-reportable quantities, such as alpha activity, are calculated and automatic quality checks are performed.

PNNL is developing an operating procedure for the SuperHENC, a mobile transuranic waste assay system.

Development of Assay Techniques for Standard Waste Boxes and Unique Burial Boxes

A standard waste box counting system was developed and qualified to estimate the amount of plutonium leaving PFP as equipment contamination, an essential part of a for special nuclear material.

For Fluor Hanford's PFP Closure Project, PNNL developed and qualified a standard waste box counting system to provide estimates of plutonium leaving the complex as contamination on equipment. These measurements are essential to maintain an accurate accounting of all special nuclear material, and the accuracy requirements are very rigorous. During the box qualifying process, techniques were used to provide the best assay value for both gamma and neutron measurement systems. Using a neutron add-a-source technique allowed correction factors to be generated and applied to the neutron assay result.



In FY04, PNNL qualified a system that counts gamma and neutron radiation in standard waste boxes, an essential part of accounting for all special nuclear material leaving PFP.

Hydrogenous materials in the waste can cause the neutron assay result to be considerably underestimated. This technique is used to correct for the matrix materials effects. A strong cesium-137 gamma source was used to provide transmission correction factors for correcting the gamma signatures emitted from the waste boxes. Combining the results of the two techniques provides the best assay values afforded by each assay system and also reduces the total measurement uncertainty.

Batch Contact Tests of Alternative Approaches to Remove Technetium from Waste Streams

To support the Liquid Effluent Retention Facility, four resins were tested to remove technetium-99 from wastewater.

Fluor Hanford is investigating treatment methods for removing technetium-99 from wastewater currently stored in the Liquid Effluent Retention Facility (Basin 44). The baseline treatment technology uses fixed-bed ion-exchange columns to selectively adsorb technetium-99 from a filtered reverse osmosis process stream and prepare the stream for secondary treatment. PNNL performed batch contact tests of four candidate resin systems to remove technetium-99 from a wastewater stream provided by the 222-S Laboratory. Besides measuring the ability of each resin to adsorb technetium-99, PNNL also measured particle size distributions. Three commercial anion ion-exchange resins and one experimental resin derived from Self-Assembled Monolayers on Mesoporous Supports (SAMMS) were tested. The tests showed that conventional ion exchange resins would perform better than the SAMMS material, but the materials were less effective than first hoped. The test results are summarized in a letter report to Fluor Hanford.

Batch contact tests of resin systems were performed to evaluate each resin's ability to remove technetium-99 from a waste stream.

Groundwater Remediation

Characterization of Systems

PNNL developed tools for Fluor Hanford to facilitate user access to data and provide information. These tools included the following:

PNNL developed the Hanford Borehole Geologic Information System to facilitate user access to data.

- A prototype graphical user interface for the Features, Events, and Processes database
- Conceptual models and input data packages for the 2004 Composite Analysis
- Web-based borehole geologic information system, a database of geologic contacts for the Central Plateau of the Hanford Site, and a Hanford Site standard for presentation of geologic data
- Methods to estimate hydraulic properties used in flow and transport models from particle-size data, and a database of contaminant distribution coefficients (K_ds) used in transport models.

Conceptual models and input data packages were developed for the Composite Analysis.

Evaluation of Remedial Alternatives at 100-NR-2

Fluor Hanford is working to evaluate remedial alternatives to reach a final record of decision for the 100-NR-2 groundwater operable unit. The interim record of decision to pump and treat groundwater is recognized as ineffective in treating strontium-90 in groundwater at the 100-N Area. PNNL is investigating two remedial alternatives: phytoremediation and apatite sequestration. Phytoremediation involves using plants to reduce the flux of strontium-90 to the Columbia River in the area near N Springs. During FY04, PNNL investigated the feasibility of phytoremediation in the laboratory. Apatite sequestration involves formation of the mineral apatite in the aquifer at the 100-N Area by injecting fluids through conventional groundwater wells. During FY04, PNNL and Sandia National Laboratories evaluated and proved that apatite precipitation occurs at the laboratory scale. Both of these evaluations are proceeding during FY05.

The feasibility of phytoremediation of strontium-90 was investigated.

Fluor Hanford is working to evaluate remedial alternatives to reach a final record of decision for the 100-NR-2 groundwater operable unit.

Wells were installed in the 100-N Area to help evaluate groundwater remediation alternatives.

River tubes were installed near N Springs to characterize discharges of contaminants.

Coyote willows were studied as one alternative to remove strontium from the groundwater under the 100-N Area.

Waste Site Remedial Action

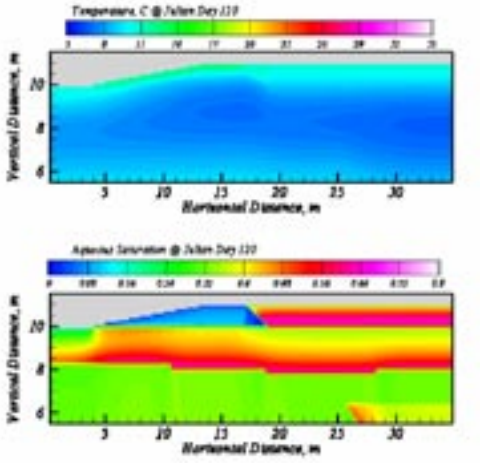
Evaluation of Carbon Tetrachloride

Fluor Hanford is conducting a remedial investigation/feasibility study of carbon tetrachloride contamination in the 200 West Area. As part of this study, PNNL evaluated and updated the conceptual and numerical models for carbon tetrachloride. In addition, PNNL applied the improved numerical model to evaluate the subsurface distribution of carbon tetrachloride and evaluate remediation alternatives.

The conceptual and numerical models of carbon tetrachloride were evaluated and updated.

U Plant Area Surface Barrier Modeling

Fluor Hanford is working to remediate U Plant area waste sites to reduce or eliminate risks of uranium and technetium-99 to human health and the environment. To support this effort, PNNL developed a new surface barrier design and optimization tool, based on the PNNL-developed Subsurface Transport Over Multiple Phases (STOMP) code. PNNL continues to support Fluor Hanford with parameterization and application of the model, as well as interpretation of the results. As part of the surface barrier modeling, PNNL has assisted Fluor Hanford in interpreting data from the borrow source area, where material will be mined for surface barrier construction. In addition, PNNL has conducted simulations of contaminant transport from hypothetically grouted residual wastes in the 221-U Canyon facility.



To support Fluor Hanford's remediation of U Plant waste sites, PNNL developed a new surface barrier design and optimization tool, based on the STOMP code.

BC Cribs and Trenches Fate and Transport Modeling

In support of Fluor Hanford's investigation of accelerating closure of the BC cribs and trenches, PNNL performed numerical simulations to evaluate the fate and transport of contaminants in no action and surface barrier alternatives. The simulations were performed using PNNL-developed conceptual and numerical models of flow and contaminant transport through the vadose zone at the BC cribs and trenches. The conceptual and numerical models were updated based on knowledge and methods developed as part of Vadose Zone Transport Field Studies.

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