

Environmental Solutions

FY05

PNNL Contributions to Fluor Hanford



Fluor Hanford

**Pacific Northwest
National Laboratory**

Operated by Battelle for the
U.S. Department of Energy

PNNL-15641

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Executive Summary

PNNL Supports Solving Nuclear Waste Cleanup Challenges

In fiscal year 2005, Pacific Northwest National Laboratory provided the scientific and technological solutions requested by Fluor Hanford, Inc. for cleaning up the Hanford Site, a former plutonium production complex in southeastern Washington. Fluor Hanford has responsibility for the overarching Hanford Site cleanup contract.



Pacific Northwest National Laboratory researchers are providing support to Fluor Hanford staff, who are remediating waste sites and protecting the Columbia River.

Demolishing Plutonium-Contaminated Buildings Without Harming the Environment or Public

Researchers helped prepare for the demolition of facilities contaminated with plutonium and other hazardous materials. By using advanced computer models combined with expertise in atmospheric dispersion, the team showed that some alternatives were more effective in controlling the spread of contamination.

To locate plutonium in ductwork, the Laboratory's researchers designed, built, and are testing sensor-packed robots. The information obtained will be used to develop safe, effective demolition plans for this complicated steel ductwork.

To protect Fluor Hanford workers, the public, and the environment during Hanford Site cleanup, PNNL provided carefully researched answers grounded in real-world experience.

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PNNL's researchers worked as part of the Fluor Hanford Nuclear Safety Team to determine the hazards from potential accidents and to find ways to mitigate these hazards.

Some wastes simply don't conform to conventional standards. Pacific Northwest National Laboratory engineers and scientists are using their experience with industrial engineering and nuclear materials to advise on the type of equipment and space needed to dismantle these highly contaminated, nonconforming wastes, which vary from concrete boxes the size of a small trailer to open paint cans filled with radioactive waste.

Moving Radioactive Sludge Away from the River and Then Disposing of the Sludge

Pacific Northwest National Laboratory's researchers provided answers needed to safely encapsulate radioactive sludge from two nuclear storage basins. This work moves the sludge away from the Columbia River. In fiscal year 2005, researchers developed a system that allows workers to easily calculate and control the optimum amount of waste encapsulated in each drum.

In addition, the team provided the experimentation and analysis to make sure that no water would separate from the waste after it was encapsulated.

Stopping Radionuclides from Reaching the River

The Laboratory helped find ways to detect, track, and stop radionuclides in the soil and groundwater. In one innovative method, microbes in the soil chew apart a specific cocktail of minerals creating apatite, a common mineral. The apatite traps and holds radioactive strontium moving past in the groundwater, protecting the river.

To address technetium, another radionuclide of concern, Pacific Northwest National Laboratory's researchers used field research and computer models to determine where the technetium was and where it was headed. The computer models were instrumental in accurately communicating the information to Fluor Hanford.

Protecting Workers and the Environment During Cleanup

In cleaning up the Hanford Site, new plans present new challenges. For example, Pacific Northwest National Laboratory's researchers worked as part of the Fluor Hanford Nuclear Safety Team to determine the hazards from potential accidents – from earthquakes to pump failures – and to find ways to mitigate these hazards. By including science and innovation as part of the process, safety challenges can be anticipated and successfully handled when they appear.

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Introduction

Fluor Hanford, Inc. is the Project Hanford Management Contract Prime Contractor for the U.S. Department of Energy's Richland Operations Office at the Hanford Site. Fluor Hanford staff are deactivating and demolishing buildings such as those once used to process plutonium. They are transporting transuranic waste to the Waste Isolation Pilot Plant in New Mexico, the world's first underground repository licensed to safely and permanently dispose of transuranic waste. Fluor Hanford staff are also remediating the K Basins, two spent nuclear fuel storage pools near the Columbia River, a vital resource in the Pacific Northwest.

Pacific Northwest National Laboratory is supporting Fluor Hanford in solving problems and finding answers to important questions that affect safety, costs, and schedule.

Highlights of the Laboratory's support to Fluor Hanford for FY05 are summarized in this booklet. Work performed 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.

Spent Nuclear Fuel Sludge

Built in the early 1950s, the K-East (KE) and K-West (KW) Basins were used to store irradiated fuel before processing. Each leak-prone K Basin is approximately 67 feet wide, 125 feet long, and about 21 feet deep, filled with approximately 1.2 million gallons of water and enclosed in a steel building. The basins contain side areas, such as the North Load Out Pit, to support specialized activities.

PNNL researchers provided scientific and technical support to Fluor Hanford for removing and disposing of radioactive sludge from the K Basins.



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Radioactive fuel was stored in the Hanford Site's K Basins, located near the Columbia River. Now, with the fuel removed, each basin still contains radioactive sludge, some equipment, and water.

Irradiated fuel from the N Reactor was placed in the basins, for what was intended to be short-duration storage. When Hanford's fuel reprocessing facility closed in 1989, the fuel remained in the basins. Fluor Hanford staff are removing the smaller and corroded fuel from the leak-prone basins and placing it into dry storage.

With scientific support from PNNL, the Fluor Hanford team is removing and disposing of 50 cubic meters of sludge, accumulated on the bottom of the basins. The highly radioactive sludge consists of degraded fuel, debris, silt, sand, and other materials. This sludge must be removed and disposed to prevent the possibility of sludge-contaminated water leaking into the nearby Columbia River.

The sludge in the KE Basin is being pumped into containers and transferred via a hose-in-hose system to the KW Basin, where along with the KW sludge, it will be pumped to another facility to be treated, assayed, and grouted in drums. The basins, which will contain some remaining materials that cannot be effectively retrieved, will be partially filled with grout, cut apart, and transported to a disposal site far from the river.

Optimizing the Volume of Waste Grouted in Each Drum

Scientists at PNNL provided innovative modeling and calculations to assist Fluor Hanford operators in adding the proper amount of sludge from the KE Basin North Load Out Pit with grout-forming materials inside a 55-gallon drum. The goal is to add the appropriate amount of sludge to approach – but not exceed – the determined radiation dose at the drum's surface. Adding too much waste will prevent the drum from being disposed as planned; too little, and the resources and storage space are wasted.

To determine the right amount of sludge to add to each drum, the original plan was to remove grab samples from the tank, analyze the samples, and determine the amount of waste that could be added. However, this method would be slow and expose staff to significant levels of radiation. Instead, PNNL



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Fluor Hanford operators prepare to mix sludge with grout-forming materials in drums. PNNL researchers made this process efficient and reliable by providing a control process that the operators could use to determine the right amount of waste to add to each drum.

With scientific support from PNNL, the Fluor Hanford team is removing and disposing of 50 cubic meters of sludge from the bottom of the spent fuel basins.

A process was developed that accurately determines the optimum amount of sludge to add to the grout-forming materials inside the drums.

PNNL scientists conducted bench- and process-scale tests to make sure that the grout process would produce an acceptable waste form.

researchers and Fluor Hanford staff modified the system to provide a safer way to obtain the information. First, a radiation monitor was added on a recirculating loop. As sludge moves through the loop during mixing, the monitor provides a reading on the radiation level. To use this reading effectively, PNNL researchers used complex models to calculate the correlation between the radiation measurements and the isotopic concentration in the waste slurry.

With this capability Fluor Hanford operators began grouting drums in October 2005 with predictable and consistent results.

Keeping Grouted Sludge Water-Free

Liquid is not allowed in the cured grouted sludge drums. For grouts based on portland cement materials, such as the KE North Load Out Pit, liquid is manifested as “bleed water” that remains after the completion of cement curing. PNNL scientists conducted tests and examined results for Fluor Hanford to determine if the grout process, using bentonite clay to absorb bleed water, would produce an acceptable waste form.

Results from bench- and process-scale testing showed that the grouted products should not produce liquid if the process is controlled under the stated parameters, even for sludges that are nearly 100% water.

Each batch of waste to be grouted consists of 100 liters combined of sludge and additional water. The bench-scale tests showed that up to 115 liters of water with no sludge solids could be tolerated at the lowest controlled quantities of grout-forming solids before free liquid could arise. The quantity of excess water needed to reach the 115-liter threshold is three times the process control performance of the grout preparation apparatus.

The PNNL researchers further showed that the actual controls for weighing the grout components are better than the design limits, especially for bentonite, and would allow at least 123 liters of water to be added to the grout drum before free liquid would appear. This provides intrinsic tolerance that is more than four times the process control performance.

Bench-scale tests conducted by the PNNL research team showed that the marginal yield of free liquid produced for saturated grouts (i.e., grouts at the threshold of producing free liquid) was only about 30% of the excess water being added. This behavior of the bentonite grout minimizes the effects of any inadvertent overdose of make-up water in the grouted sludge preparation.

Overall, the PNNL research team showed that the process controls, equipment design, and knowledge of the behavior of the grouted sludge waste form could produce grout from the KE North Load Out Pit waste without generating free liquid.

Deactivating and Decommissioning

Modeling Radionuclide Deposition and Dispersion to Support Demolition

Researchers at PNNL used sophisticated models to predict how various demolition alternatives would disperse radioactive materials. The researchers defined relationships between the amount of residual contamination in the buildings, and the air and soil concentrations that may be generated by demolition. This information will help Fluor Hanford managers control potential contamination within established limits.

To provide this information, the PNNL analysts conducted atmospheric dispersion modeling. Because the “demolition ready” radiological condition of the Plutonium Finishing Plant complex has not been specified, the team took a parametric approach to the source term. They considered a range of radiological conditions of the PFP buildings at the time of demolition. Release fractions for open-air demolition approaches were evaluated that spanned potential emission rates.

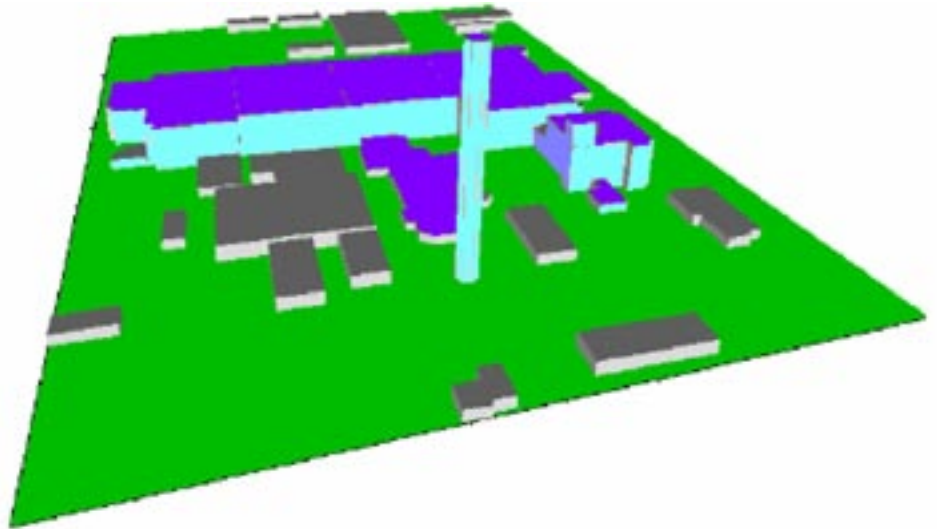
To support demolition plans, PNNL researchers modeled the atmospheric dispersion of contamination.



Deactivating and dismantling Plutonium Finishing Plant systems and structures eliminates significant hazards to workers, the public, and the environment. In addition, deactivating and dismantling minimizes long-term surveillance and maintenance risks and costs.

Local air dispersion patterns were considered. These patterns are created by the interactions of the PFP and surrounding facilities with wind and other meteorological phenomena. In addition, the researchers used hourly meteorological data collected over 1 year to examine the effects of wind speed, direction, and stability on projected concentrations. Air and soil concentration estimates were made for the daily combinations of weather conditions by modeling each hour during the day. To be conservative, it was assumed that precipitation did not occur during demolition activities. The resulting 24-hour air concentrations and total annual deposition patterns provided the basis for defining the maximum acceptable residual contamination.

The controlling factor in demolition planning is likely to be the cumulative deposition of alpha-emitting radionuclides on soils and surfaces near the buildings, rather than the worker exposure to contaminated air. The relatively low concentrations of radioactive materials in air, protracted over an extended period of time, result in a relatively high cumulative deposition. The need to limit the releases to minimize the cumulative deposition still allows manageable amounts of plutonium contamination within the buildings to be demolished.



Modeling dispersion around buildings is highly complex, given building wake effects, meteorological conditions, and other phenomena. PNNL researchers provided simple and complex models of the dispersion around the PFP complex. In this simple model, structures in blue are scheduled for demolition.

Testing Robots to Characterize Plutonium in Ducts

As part of PNNL's ongoing effort to assist Fluor Hanford in decommissioning plutonium-contaminated ductwork, characterization tools and training mockups were developed for activities at the 291-Z Air Filter and Exhaust Stack Facility and 232-Z Incinerator Building.

For 291-Z remediation, PNNL researchers provided remote characterization tools and built a training mockup. The tools included the Variable Geometry Tracked Vehicle that was modified to carry a radiation sensor that characterized the inside of the ducting, providing both radiation readings and video images.

In addition, PNNL researchers provided similar support for the 232-Z duct remediation effort. The engineers at PNNL developed a robotic video inspection device to move through these smaller ducts. The camera is manipulated inside the ducting using rare earth magnets built into the camera sled and into handles used by the operators on the outside of the ducting.



PNNL researchers developed a simple robot and camera system (left) and modified the Variable Geometry Tracked Vehicle (right) to carry a radiation sensor through the ductwork and meet safety requirements.

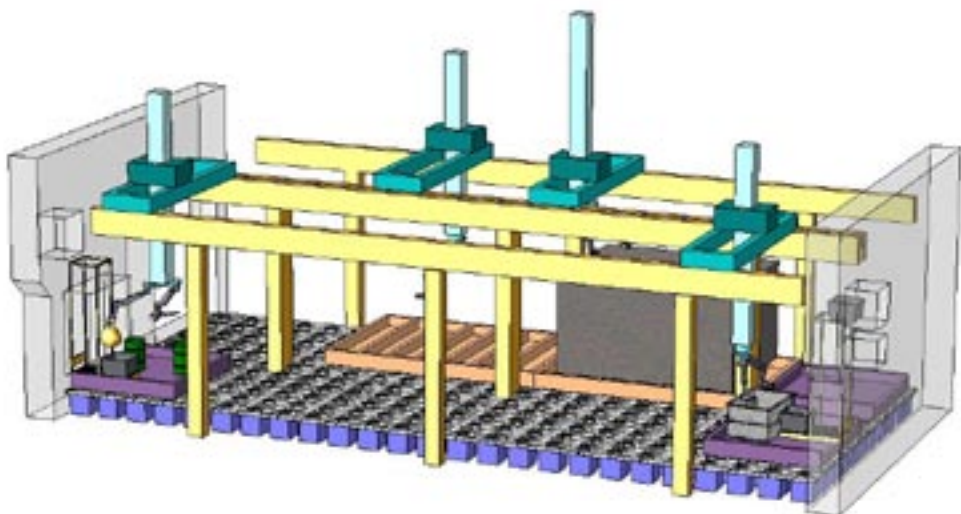
Framing the Technical Issues for Dismantling Very Large and Very Small Items

PNNL researchers are helping Fluor Hanford staff prepare for dismantling and disposing of highly radioactive wastes in various sizes. One example is concrete boxes from PFP that weigh several tons and are more than 20 feet long. These boxes were used to store pumps and other contaminated equipment. Another example is paint cans filled with highly contaminated materials. The radioactivity levels, mostly alpha radiation from plutonium exposure, require that all of the work be done remotely. The varying size of the waste means that a number of different functions, from removing paint can lids to breaking apart concrete, must be available.

Using their experience with robotics and nuclear materials, PNNL researchers advised on the type of equipment and space needed to handle these wastes. The team also worked to develop the system with a focus on maintenance, replacement, and decommissioning. This is an ongoing project for PNNL and Fluor Hanford.

PNNL provided remote characterization tools to obtain radiation readings and video images of contaminated ductwork.

PNNL researchers advised on equipment and space for handling radioactive materials of varying sizes.



Using information from PNNL researchers, the configuration of space and equipment to handle highly radioactive materials of varying size is being explored.

Groundwater Remediation

Testing a Microbe-Delivered Method to Sequester Strontium-90

PNNL scientists investigated sequestering strontium-90 in the Hanford groundwater by delivering a solution that microbes slowly degrade into apatite. The apatite forms a filter in the soil that traps the strontium. The current pump-and-treat system only removes about 0.2 curies a year, 10 times less than is decaying naturally.

PNNL researchers conducted laboratory-scale studies to quantify the sequestration processes. They determined that strontium-90 is initially held by ion exchange, but over 6 to 20 weeks, the strontium is more permanently held, effectively preventing it from moving with groundwater.

PNNL researchers also showed that apatite precipitation occurred within 75 to 200 hours, depending on the oxygen level in the soil.

Laboratory tests showed that without apatite, 90% of the strontium-90 could be removed from the sediment by ion exchange. With apatite, only 45% of the strontium-90 could be removed after 6 weeks, and this slowly decreased to 32% by 24 weeks. Researchers believe this may be caused by slow strontium-90 incorporation into apatite.

To minimize strontium-90 mobilization during barrier emplacement, a sequence of solution injections from low to high concentration is needed. Mobilization is minimal with the low-concentration injection. Then, a high-concentration

The apatite technology was evaluated to determine if it would immobilize strontium-90 for 300 years.

injection can be added with minimal strontium-90 desorption. This sequential injection scheme was successful at the laboratory scale and will be deployed in a pilot-scale test in summer 2006.

Characterizing the Soil to Understand Where Radionuclides Migrate

Determining the fate and transport of radionuclides underground can involve difficult-to-obtain samples and expensive chemical analyses. Researchers at PNNL used core samples and other data to determine if geochemical parameters could be estimated from more readily obtained measurements. The researchers collected core and grab samples at the Integrated Disposal Facility open excavation and Pit 30, then measured the sample properties, including total oxide composition, particle size, total and inorganic carbon content, cation exchange capacity, and hydrous oxide content. These data are being used to help estimate geochemical transport parameters from particle-size and other physical measurements.



Consistent data, parameters, and conceptual models were developed to better understand and remediate radionuclides in soil.



Researchers at PNNL used core samples to help resolve the gravel issue around waste migration.

PNNL scientists carefully take core and grab samples from Pit 30. The results from analyzing these samples will help Fluor Hanford staff determine if geochemical parameters can be reliably estimated from easily measured physical parameters.

Information on technetium-99 transport will be used to make decisions about groundwater cleanup.

The PNNL team also characterized the Pit 30 sediments to provide data that would be “standards” for future geochemical studies and improve the database on the gravel K_d [distribution coefficient] correction issue. The issue occurs because previous data on the soil below Hanford disposal sites did not always account for material greater than 2 millimeters in size.

Additional information on this work can be found at <http://www.hanford.gov/cp/gpp/modeling/cos.cfm>.

Modeling Soil Transport of Technetium-99

Fluor Hanford staff are developing an effective remediation plan and evaluating remedial options for the BC cribs and trenches using a detailed analysis of fate and transport provided by PNNL researchers. These cribs and trenches are believed to have received about 30 million gallons of waste containing an estimated 400 curies of technetium-99, as well as large quantities of nitrate and uranium-238.

Because the subsurface area under the cribs and trenches is very heterogeneous, PNNL researchers completed field research and laboratory analyses to develop high-resolution models that reflect these small-scale differences. The field work included collecting soil samples and high-resolution neutron moisture logs. By developing relationships between the properties of the samples and logs, the

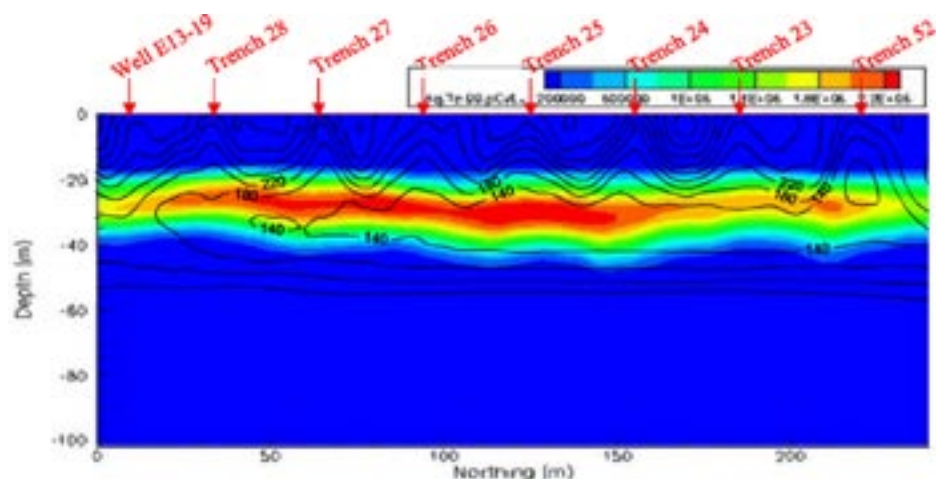


Concerns are being raised about the possible leaching of radioactive materials from Hanford trenches to the groundwater and eventually the Columbia River.

researchers were able to obtain detailed vertical information on the subsurface. In addition, the team was able to incorporate detailed knowledge on how contaminants and groundwater move horizontally at the well-characterized 299-E24-111 test site to extend the small-scale heterogeneities at 216-B-26 trench to three dimensions.

All this information was then fed into a computer simulator to predict how contamination would move through the subsurface under different remediation scenarios. Computer simulations show that small-scale stratigraphic differences strongly affect flow in the vadose zone with much of the discharged water and contaminants spreading laterally in the shallow subsurface before migrating downward. Simulations are in good agreement with field observations. These scenarios were used to predict the effects of cleanup options over 22 acres and for thousands of years.

Fluor Hanford staff will use this information to make decisions about groundwater cleanup.



Researchers at PNNL calculated distributions of aqueous technetium-99 (color contours) and compared the information to noninvasive geophysical measurements of electrical conductivity (black contour lines).

Worker and Environmental Safety

Developing a Safety Basis for K Basin Sludge Disposal

PNNL researchers are leading the team providing hazards and accident analyses to develop a safety basis for the K Basin sludge disposal project. The project includes removing sludge from the K Basins using a complex overland pumping scheme, underwater settling tanks, assay, and packaging in grouted drums.

For K Basin sludge disposal, PNNL is providing hazard and accident analyses.

During FY05 the PNNL team, coordinating with Fluor Hanford staff, made major contributions to the sludge treatment process system design, and developed information needed for criticality and additional safety analyses, as well as regulatory reviews. For example, PNNL researchers provided answers on hazards and subsequent risk management. This is an ongoing effort.

In addition, PNNL conducted hardness measurements on irradiated uranium fuel. With this information, the PNNL research team developed a mechanical simulant to use in pump tests. Based on the results, Fluor Hanford installed redundant pumps to protect against problems caused by erosion of the pumps' components.

The PNNL team was selected for this task because of past successful experience with safety analysis and production of safety documentation that meets client and regulatory requirements. The team members consisted of accomplished staff experienced with hazard and accident analysis on nuclear, oil, medical, chemical, and process systems.

Determining Safety Limits for Aluminum in Grout

To immobilize residual equipment and debris in the KE Basin, Fluor Hanford will partially fill the basin with grout. The grout is alkaline and will corrode the aluminum in some of the debris, generating hydrogen gas. So, PNNL researchers performed analyses to determine the amount of hydrogen that will be generated during grouting and identified measures to avoid a hydrogen-based accident.

Based on the review of experimental and analytical studies, the researchers concluded that the likelihood of generating a flammable mixture of hydrogen with grout is low, but not zero. The PNNL team developed recommendations to control the distribution of aluminum inventory on the basin floor and minimize temperatures during the grouting operations:

- The residual aluminum metal inventory in the basin, especially the fuel canisters, should not be stacked on top of one another to prevent concentrating the aluminum metal inventory over a small surface area.



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Radioactive sludge, shown on the left, at the bottom of one of the Hanford Site's spent nuclear fuel storage basins presents cleanup and safety challenges.

PNNL researchers performed analyses to determine the amount of hydrogen that will be generated during grouting and identified measures to avoid a hydrogen-based accident.

- The temperature of the grout should be kept under 194°F during pouring and for at least 3 hours after the aluminum metal has been covered. Lower temperatures result in less hydrogen being generated.
- The basin water temperature should be less than 140°F for at least 3 hours after a grout pour is interrupted and the aluminum metal has not been completely covered. This minimizes the reaction of the uncovered aluminum metal with calcium hydroxide.
- The basin water should not be removed while the grout is being poured. This is not necessary if the water removal system is appropriately vented or after the aluminum metal has been covered with grout for at least 3 hours.



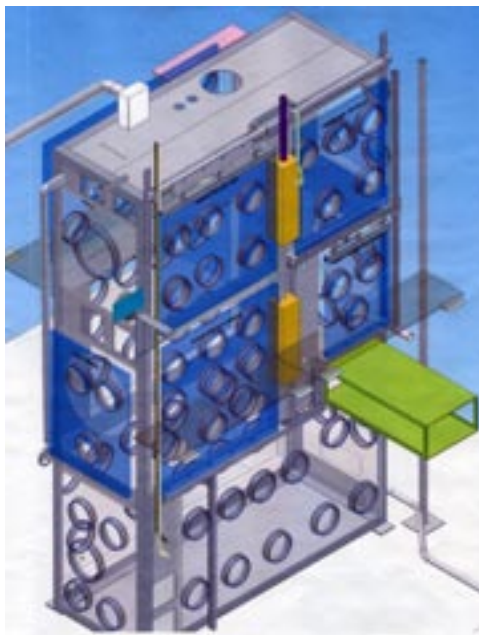
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While some debris will be removed from the K Basins, empty fuel canisters, identification tags, and other material containing aluminum will be grouted in place, then later removed as a large monolith.

Developing Safe Limits to Dispose of Glovebox Decontamination Materials

To assist Fluor Hanford in decontaminating more than 200 gloveboxes, used for chemical operations at the Plutonium Finishing Plant, PNNL researchers provided a technical basis for safely disposing of proposed chemical decontamination agents, cloth rags, and vacuum cleaner filters used to remove the agents. This waste is a concern because the chemicals are generally very reactive, and under certain conditions, could develop self-heating reactions, potentially leading to fires inside the waste drums.

PNNL provided guidance for safely decontaminating gloveboxes, where most of the Plutonium Finishing Plant's chemical operations took place.



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Decontaminating gloveboxes at PFP can be very complex, as this schematic of the HA-9A box shows. PNNL researchers helped make the process safer by providing data on the reactivity of chemical agents and other materials used to decontaminate these large, complex systems.

The PNNL research team used laboratory thermoanalytical methods and thermal modeling of the waste packages to evaluate the behavior of combinations of decontamination chemicals and waste removal/packaging materials. The PNNL team translated the analytical results into safe operating procedures. These procedures will protect the environment and workers from the dangers inherent in self-heating waste drums.

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