Environmental Solutions

A Summary of Contributions for FY04

PNNL-15094

PNNL Contributions to CH2M HILL Hanford Group, Inc.

CH2MHILL Hanford Group Inc

Pacific Northwest National Laboratory
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Executive Summary

Partnership Supports Tank Waste Remediation

In 2004, Pacific Northwest National Laboratory provided analyses, reviews, testing, and new tools to assist CH2M HILL Hanford Group, Inc. in accomplishing their River Protection Project objectives. These objectives are safe storage, retrieval, and treatment of radioactive waste from Hanford's tanks, closure of tanks, and disposal of treated wastes.

While certain portions of the waste in Hanford's 177 tanks will be vitrified at the Waste Treatment Plant, DOE has tasked CH2M HILL with evaluating supplemental treatment processes to immobilize part of the less radioactive or low-activity waste. CH2M HILL and their contractor AMEC Earth and Environmental Inc. are conducting tests to determine if bulk vitrification can be used to supplement the treatment capacity of the Waste Treatment Plant. This process creates large glass blocks, greater than 20 feet in length. In 2004, PNNL developed a simulated waste and used it in tests to evaluate and refine the glass formula and the bulk vitrification process.

After the bulk of the waste is removed from the tanks, CH2M HILL prepares the tanks for closure, marking the end of their time as storage vessels. In 2004, PNNL assisted CH2M HILL in assessing the fate of the small fraction of remaining waste. Using residual waste samples and more than 40 years of experience with Hanford's geochemistry, PNNL developed computer models that predicted how and under what conditions contaminants

In supporting CH2M HILL's challenging mission to manage the waste in Hanford's tanks, PNNL is helping to protect the sustainability and health of the Columbia River.

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Tests were conducted to determine if bulk vitrification can be used to supplement the Waste Treatment Plant.

would be released from the residual waste. In addition, PNNL is conducting detailed studies on the soil under the tanks to assess waste migration pathways if a leak occurred during retrieval.

Vitrified low-activity wastes and other solid wastes will be disposed at the Integrated Disposal Facility, a landfill being built in Hanford's 200 East Area. In 2004, PNNL provided data on the geology, hydrology, and geochemistry of the area. The Laboratory also tested waste glasses that will be in the facility, looking at how technetium and other contaminants would behave when the glass is placed in a disposal environment.

In retrieving the waste from the tanks, as well as day-to-day operations, chemical vapors from the tanks are a concern. Supporting CH2M HILL's efforts to protect its workers, PNNL provided data and analyses on the vapors emitted through the ventilation systems on each tank and how the vapors were dispersed. In addition, PNNL tested the accuracy, precision, and response time of several commercially available industrial hygiene field instruments to potentially be used by tank farm workers.

To help CH2M HILL achieve the accelerated schedule for retrieving double-shell tank waste, a new software tool called the Compatibility Assessment Automation Tool, or CAAT for short, was developed by PNNL. Using existing data within the Tank Waste Information Network System, or TWINS, this software simplifies solving a number of calculations regarding tank waste transfers.

Because the River Protection Project schedule requires waste be held in the double-shell tanks beyond their original design life, CH2M HILL is evaluating the tanks to ensure they are fit for duty. In support of this evaluation, PNNL analyzed the tanks' interiors and evaluated the effects of temperature changes, tank waste, and earthquakes.

Contents

Processing the Waste

Supplemental Treatment with Bulk Vitrification

In support of CH2M HILL's bulk vitrification projects, Pacific Northwest National Laboratory provided waste feed modeling, waste feed simulant development, waste form qualification planning, waste form performance enhancement, and waste form characterization/modeling to support risk assessment activities.

Major FY04 accomplishments included the following:

- Conducted computer modeling to generate a baseline simulant formulation for the retrieved tank S-109 waste that will be used in the Demonstration Bulk Vitrification System.
- Worked with U.S. Department of Energy, CH2M HILL, and regulators to generate a set of waste form qualification objectives and have established an initial strategy that specifies how all objectives will be met.

A PNNL researcher prepares to use a centrifugation technique to remove the pore-water extractable rhenium from castable refractory samples. Rhenium is a non radioactive surrogate for technetium in laboratory studies.

- Developed a method to measure the soluble fraction of technetium that is deposited in the bulk vitrification refractory and used this method to guide efforts to reduce these deposits.
- Developed and tested a two-dimensional waste form release model that can predict the release of contaminants from the complicated bulk vitrification waste package. This model will be used as part of CH2M HILL's bulk vitrification risk assessment activities.
- Characterized the glass product produced from tank S-109 simulant and all bulk vitrification waste form package components necessary to support CH2M HILL's bulk vitrification risk assessment activities.

PNNL provided scientific studies and technical assistance to improve the process and product from bulk vitrification of low-activity waste from Hanford's underground tanks.

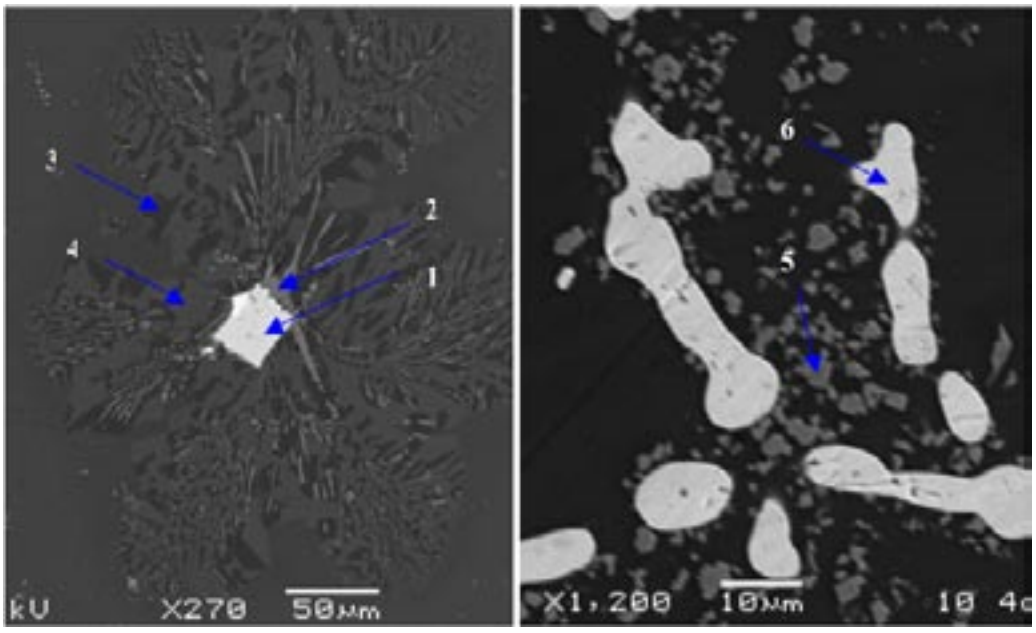
Technical Assistance to AMEC Earth and Environmental Inc., Bulk Vitrification Vendor

PNNL provided extensive technical assistance to CH2M HILL's bulk vitrification vendor, AMEC Earth and Environmental Inc.

In support of CH2M HILL's bulk vitrification contractor, PNNL conducted tests to evaluate and refine the glass formula and the bulk vitrification process.

Laboratory-Scale Tests

PNNL conducted laboratory-scale tests using crucible melts to thoroughly evaluate AMEC's baseline glass formulation. A series of glasses were prepared and subjected to a battery of tests to confirm their contaminant release performance. These tests included toxicity characteristic leaching procedure, product consistency test, and vapor hydration test at the Applied Process Engineering Laboratory. In addition, phase identification tests were conducted using optical microscopy, scanning electron microscopy, and x-ray diffraction.



Crystalline phases were identified in AMP1-03-SC glass (1-Baddeleyite; 2-Sodium-zirconium silicate; 3-nepheline; 4-combeite, 5-spinel, 6-Fe metal) using a scanning electron microscope.

Engineering-Scale Tests

In the Radiochemical Processing Laboratory, PNNL supported radioactive engineering-scale tests to evaluate the bulk vitrification process and support a technetium material balance. The glass in the engineering-scale melts was made by combining soil, small amounts of chemical additives, and simulated tank waste in a container about the size of a desk. The mixture was then heated, vitrifying the simulated waste.

At the Radiochemical Processing Laboratory, this setup was used to conduct engineering-scale bulk vitrification tests on technetium-99 spiked simulants.

Waste Form Evaluation

PNNL evaluated the waste forms created in other nonradioactive engineering-scale tests. Selected samples of the simulated waste form underwent toxicity characteristic leaching procedure and vapor hydration tests at the Applied Process Engineering Laboratory. In addition, phase identification tests were conducted using optical microscopy, scanning electron microscopy, and x-ray diffraction. These tests provided data on the consistency of the melt.

PNNL evaluated waste forms created in nonradioactive engineering scale tests.

Closing Tanks and Disposing of the Waste

Residual Tank Waste Source Release Models

PNNL is developing source release models for contaminants held in the small amount of waste that will remain in the tanks after retrieval and closure. These release models are an important component of the risk/performance assessment for single-shell tank closure performed by CH2M HILL. PNNL develops the models by laboratory testing residual waste to identify the solid phases containing the contaminants and the leachability of contaminants from those phases by water, which might enter the tank over thousands of years.

Source release models are being developed for residual tank sludge.

During FY04, testing of sludge from tanks BX-101 and AY-102, which began in prior years, was completed and documented. Samples of sludge from tanks C-203 and C-204 were received in December, and residual sludge samples from C-106 were received in April. Laboratory testing and release modeling for these sludges have been completed and documented in draft reports awaiting final reviews.

Several sludge leaching tests showed that tank waste contaminants considered to be very mobile (such as technetium, uranium, and iodine) are not always water leachable. For tank C-106, greater than 90% of these contaminants in the waste could not be leached by contact with water. Technetium from waste in tanks AY-102, C-203, and C-204 was also not readily leached with water. These results allow for more realistic risk/performance assessments, which in the past assumed high leachability for these contaminants. It has also been found that the wastes in different tanks have very different compositions and contaminant release characteristics. At some point, it may be possible to group tanks together that have similar contaminant release characteristics, but at this stage, there is no obvious trend that would allow such a grouping.

During FY04, PNNL completed its analysis of unleached sludge from tank BX-101. Several of the tests showed that some tank waste contaminants were not as leachable as previously thought.

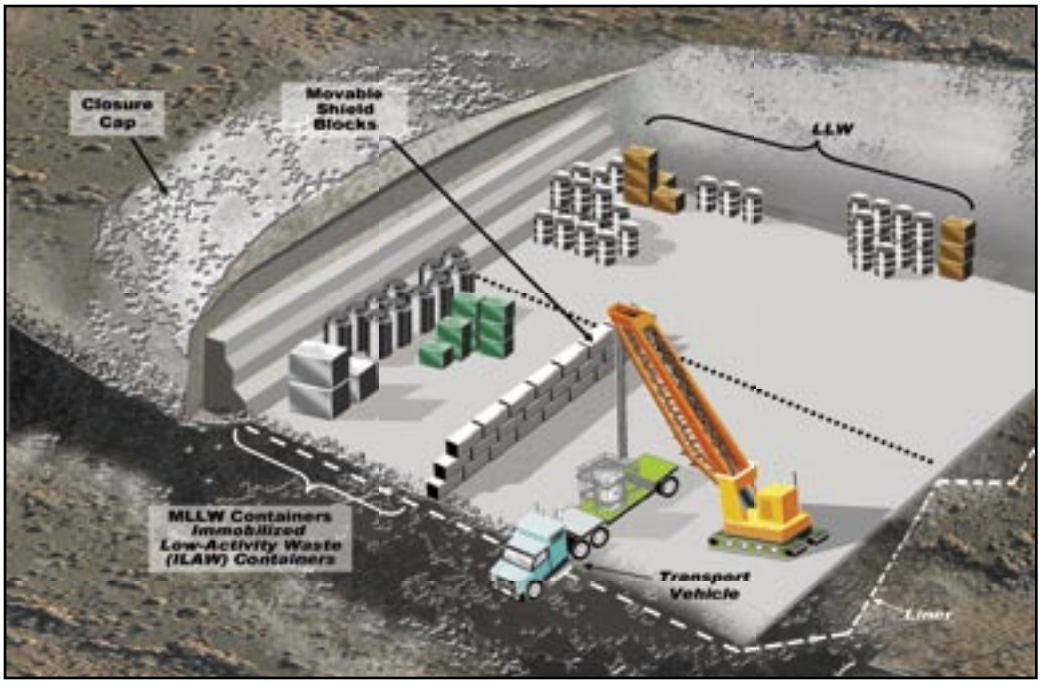
Integrated Disposal Facility Performance Assessment

PNNL provided CH2M HILL with data to support calculations of contaminant release from the Integrated Disposal Facility or IDF. In addition, PNNL has developed a multiphase flow and reactive chemical transport model called STORM that integrates the laboratory and field data collection activities to provide contaminant fluxes to the vadose zone extrapolated more than 10,000 years into the future. This model is executed at the Environmental Molecular Sciences Laboratory's supercomputer facility.

For the Integrated Disposal Facility, a complex multiphase flow and reactive chemical transport model that extrapolates contaminant fluxes to the vadose zone for more than 10,000 years was developed.

Specific accomplishments on the IDF performance assessment in FY04 include the following:

- Issued data packages for the 2005 IDF performance assessment in geology, hydrology, geochemistry, and waste form. These data packages form the technical basis for the effort by DOE to obtain consistency across the Hanford Site's assessments.
- Completed the first-ever pressurized unsaturated flow tests on immobilized low-activity waste glasses made with actual tank waste. No evidence for technetium incorporation into secondary phases was found, contrary to observations in standard water-saturated static experiments.



Provided courtesy of CH2M HILL Hanford Group

Pacific Northwest National Laboratory provided data that will be used to calculate releases from the Integrated Disposal Facility, a landfill for low-level and mixed low-level waste.

- Discovered much lower anticipated rate of recharge through the IDF, 0.1 mm/yr instead of 4.2 mm/y. This could lower contaminant release rates by as much as a factor of 50 as compared with the 2001 immobilized low-activity waste (ILAW) performance assessment.
- Reevaluated unsaturated hydraulic properties of fractured glass waste forms. The net effect is to increase water content and diffusion behavior in fractured glass, increasing calculated release rates by about a factor of 5 as compared with the 2001 ILAW performance assessment.
- Discovered lower adsorption of uranium on Hanford soil than previously expected when exposed to ILAW glass leachates loaded with sodium and carbonate produced from dissolved carbon dioxide in the air. This will increase the contribution of uranium to overall dose calculations and shorten its arrival time to a receptor.
- Adapted the STORM source-term model to handle co-disposal of multiple waste forms in the IDF including Waste Treatment Plant ILAW glass, bulk vitrification glass, and grouted secondary waste.
- Continued to monitor and collect samples from the Field Lysimeter Test Facility where two different simulated ILAW glasses are buried.

The STORM source-term model was adapted to handle co-disposal of multiple waste forms in the IDF.

Sediment Characterization

PNNL is helping CH2M HILL determine the nature and extent of vadose zone contamination and waste migration pathways from tank farm activities. This research, performed in support of the Tank Farm RCRA Corrective Action process as documeneted in the *Hanford Federal Facility Agreement and Consent Order* (part of the M-45 milestone series), is helping to assess and mitigate potential retrieval-induced leaks.

A major aspect of PNNL's research has focused on studying the thin layers of sediment left by cataclysmic floods thousands of years ago. Previous research has shown that these alternating lenses of fine-grained material have a significant impact on the mobility of subsurface contaminants by inducing horizontal migration of contaminants hundreds of meters from the tanks.

Additional FY04 accomplishments included the following:

- Common ion exchange is a key mechanism influencing the distribution of contaminants within the portion of vadose zone contacted by tank liquor. Chemical analysis of salt plume data suggest that fluid leaked from tank T-106 traveled horizontally, within the lower Hanford formation H2 unit and the two Cold Creek Formation units, at least 30 meters to the west of the T-106 tank bottom.

- Results from TX tank farm sediments indicate that three salt plumes exist in the vadose zone. The source of contamination can be traced (geochemically) to leaks from tanks TX-104, TX-105, and TX-107.
- Analysis of vadose zone samples from the A-AX Waste Management Area indicated that chloride facilitated crevice corrosion and stress corrosion cracking caused the failure of two *Resource Conservation and Recovery Act* groundwater monitoring wells. Consequently, standard operating procedure for preparation of groundwater monitoring wells includes the use of cement, instead of bentonite pellets, to backfill the annulus around the stainless steel casing in zones consisting of fine-grained or silty material.

Geochemical analysis of cores collected from the 200 Area of the Hanford Site has highlighted the effect lenses of fine-grained material have on the mobility of subsurface contaminants. Differences in the hydraulic conductivity of the fine-grained material and the overlying sand can lead to the horizontal migration of contaminants hundreds of meters from the tanks.

Protecting Tank Farm Workers

Tank Vapor Solutions Project

PNNL has provided key technical support to CH2M HILL in the areas of tank headspace sampling, emission dispersion modeling from both passive emission sources and mechanical exhauster stacks, toxicological evaluations of vapors, the development of a list of high-priority chemicals of potential concern, statistical and technical evaluation of existing and recent sampling data, and the development of supporting documents for the Industrial Hygiene Chemical Vapor Technical Basis Document. PNNL has collaborated with CH2M HILL and co-authored several documents:

- PNNL-14767, *Characterization of the Near-Field Transport and Dispersion of Vapors Released from the Headspaces of Hanford Site Underground Storage Tanks*
- PNNL-13366 Rev 1, *A Survey of Vapors in the Headspaces of Single-Shell Waste Tanks*
- PNNL-14831, *Overview of Hanford Site High-Level Waste Tank Gas and Vapor Dynamics*
- TWS04.007, *Toxicological Review Criteria for Chemicals of Potential Concern*
- TWS04.011, *Revised List of the Hanford Tank Farms Chemicals of Potential Concern.*

Sensitivity Testing for Industrial Hygiene Field Instruments

For CH2M HILL, PNNL conducted tests to evaluate the accuracy, precision, and response time of certain commercially available industrial hygiene monitors. The tests were conducted in the Chemical Chamber Test Facility. The monitors were tested with a set of dilute gases including ammonia, nitrous oxide, and a mixture of organic vapors (acetone, benzene, ethanol, hexane, toluene, and xylene). The certified gases were diluted

PNNL tested the accuracy, precision, and response times of the MIRAN along with several other monitors that may be used to detect gases emitted by Hanford's tanks.

To protect tank farm workers, chemical vapors emitted through the ventilation systems on each Hanford tank are being studied.

to concentrations that may be encountered in the outdoor environment above Hanford's underground tank farms. The challenge concentrations are near the lower limits of instrument sensitivity and response time. The performance test simulations were designed to look at how the instruments respond to changes in test gas concentrations that are similar to field conditions. The instruments evaluated are listed below by the type of challenge gas.

The accuracy, precision, and response time of several handheld vapor monitors were evaluated.

Monitor	Manufacturer	Challenge Gas
MIRAN SapphIRe XL	Thermo Environmental Instruments	Nitrous oxide and ammonia
iTX	Industrial Scientific Corporation	Ammonia
EC-P2	Manning Systems, Inc.	Ammonia
580 EZ	Thermo Environmental Instruments	Total hydrocarbons
Area-RAE	RAE Systems	Total hydrocarbons
ppb-RAB	RAE Systems	Total hydrocarbons

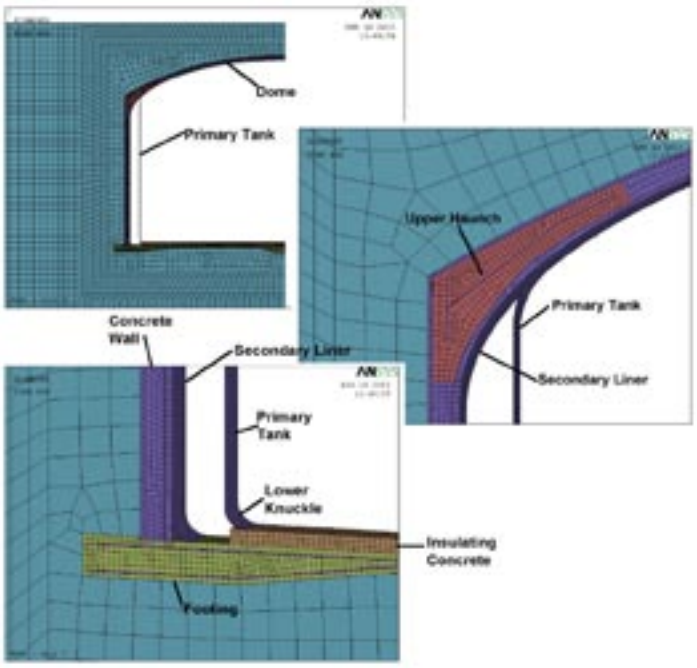
Safely Storing Tank Waste

Double-Shell Tank Thermal and Seismic Analysis

PNNL is conducting an analysis of record of the Hanford Site double-shell tanks, or DSTs, in support of the DST Integrity Program. This work will provide a structural evaluation of a representative and bounding DST and will be used as part of the documentation from CH2M HILL to the Washington State Department of Ecology to demonstrate that the DSTs are fit for continued service. This is year two of a three-year project.

Major accomplishments included the following:

Parametric Studies on Primary Liner and Insulating Concrete. PNNL completed a parametric study to evaluate the effects of various liquid levels and waste specific gravities on the tank primary liner. This information was used to help CH2M HILL define potential new liquid levels that would allow for additional waste storage in the DSTs. The team also completed a parametric study that addressed the potential effects of degraded insulating concrete on the reinforced concrete and the primary liner of the DST.

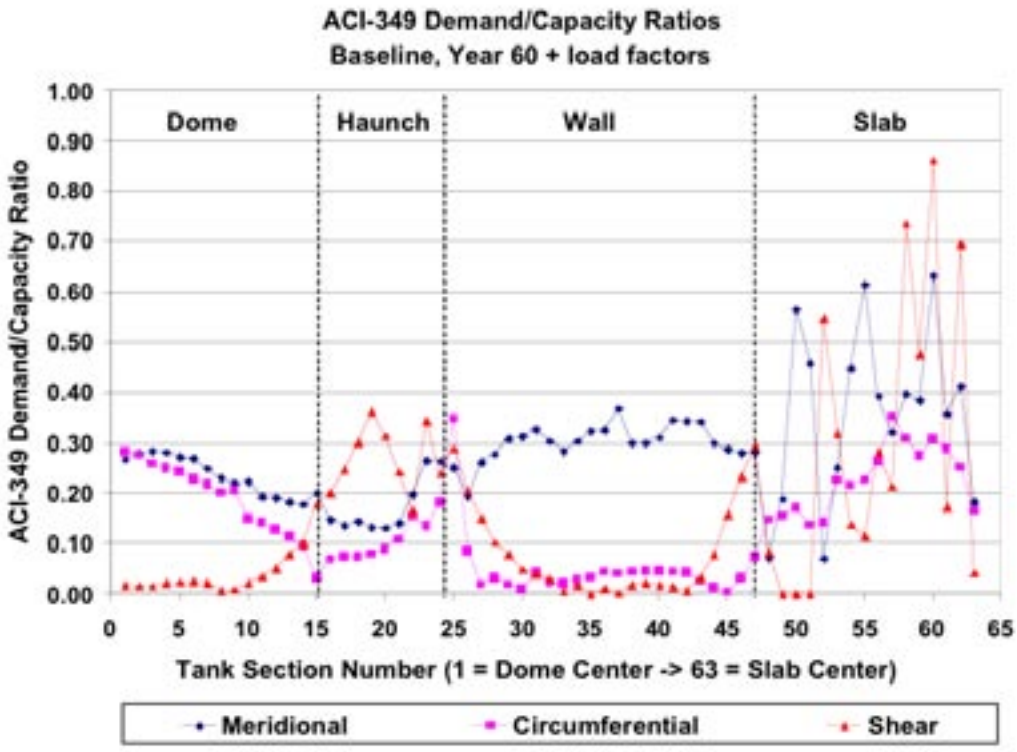


Using a computer modeling software, PNNL analyzed the thermal and operating loads for Hanford's double-shell tanks.

A parametric study that addressed the potential effects of degraded insulating concrete in double-shell tanks was completed.

Modeled Thermal Cycling and Creep. PNNL ran the bounding tank model to simulate 60 years of thermal cycling and creep. The model accounts for the effects of temperature on the properties of the reinforced concrete, thermal cycles from 50°F to 350°F, as well as the traditional dead loads and live loads. Load combinations, with the exception of the seismic, have been evaluated as well. The model results were evaluated to the ACI-349 criteria for the structural concrete and sections of the ASME Boiler and Pressure Code for the liners and bolts. The initial review of the results indicates that the tanks will pass the evaluation criteria for nearly all conditions.

Seismic Analysis. The seismic analysis of the DSTs is well underway. With help from PNNL's subcontractor M&D Professional Services, the seismic analysis methodology is using explicit time history for the input loading instead of implicit methods based upon Soil and Structure Interaction (SASI) analyses of the past. Explicit methods should be more accurate with less uncertainty.



A typical analytical result for the concrete shows the demand/capacity ratio at each cross section of the tank.

Determining Minimum Tank Wall Thickness

PNNL supported CH2M HILL on several tank projects related to determining remaining minimum wall thickness based on ultrasonic measurements. To estimate the minimum thickness for tank AY-101, modifications were made to an extreme value estimation approach initially proposed by Savannah River Site personnel. The modifications included using an alternative extreme value probability distribution that better fit the data and providing confidence bounds on the resulting extreme value estimates. This work led to participation on an expert panel workshop on raising waste levels in the DSTs.

PNNL staff also reviewed the DST Integrity Plan with particular emphasis on the suitability of using a single riser for obtaining the ultrasonic wall thickness measurements in subsequent tanks, and then extrapolating the results to cover the entire tank. The tank AY-101 methodology was appropriate in that case because considerable data were available and because four different risers were used to obtain the data.

Currently, the Laboratory is returning to the tank AY-101 measurement data to perform analyses, a single riser at a time, by taking random subsets of data and treating them as if they had been obtained in a single riser application. Differences within and between risers will be characterized in this manner and lead to a methodology for incorporating potential riser differences when only a single riser is used on a subsequent tank.

Expert Panel Review of DST Corrosion Chemistry Optimization

PNNL provided a tank integrity specialist to the Expert Panel Review of DST Corrosion Chemistry Optimization. The committee provided recommendations on the initiatives, the ultrasonic monitoring program, and a potential in situ corrosion monitoring program. As a member of this panel, the specialist helped review three initiatives regarding the tank chemistry and monitoring programs. Specifically, the staff member provided advice on the likely impacts on stress corrosion cracking if the initiatives were implemented.

PNNL provided a tank integrity specialist to the Expert Panel Review of DST Corrosion Chemistry Optimization.

Compatibility Assessment Automation Tool

The CAAT uses existing Tank Waste Information System data and automates the execution of existing capabilities.

To assist CH2M HILL in performing the long-range planning that complies with the Tank Farms Documented Safety Analysis, while also achieving the accelerated schedule for tank waste retrievals, PNNL developed the Compatibility Assessment Automation Tool, or CAAT. The CAAT uses existing Tank Waste Information Network System or TWINS data and automates the execution of existing capabilities. The capabilities assess all waste compatibility parameters including calculations of waste group assignments, calculations of time to lower flammability limit, and toxicological source term calculations. The tool can be used to assess single tank-to-tank transfers, and it can execute Hanford Tank Waste Operations Simulator (HTWOS) generated process flowsheet "Cases," which may involve over a hundred transfers extending out 5 to 10 years. CAAT dramatically speeds the acquisition of data, the execution of models, and the management of results.

The new Compatibility Assessment Automation Tool compliance report screen dramatically speeds the acquisition of data, the execution of models, and the management of results.

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