

## Department of Energy Tank Closure – An Examination of Alternative Approaches - 15390

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### ABSTRACT

Closure of single- and double-shell underground storage tanks at sites across the DOE complex poses a unique technical and regulatory challenge. Some sludge waste residues invariably remain in a tank after waste retrieval – especially in tanks with obstructions or associated piping. In addition, reducing the volume of these waste residues becomes increasingly difficult depending on the constituents and the age of the heel. This paper discusses tank cleaning and closure alternatives to allow a more efficient retrieval and characterization of the tank wastes.

The paper also examines the possible use of a tank closure strategy that is informed by risk rather than subjective criteria. Gaps in the technical foundation supporting tank retrieval and closure have traditionally resulted in a number of overly conservative assumptions in performance assessments. An alternate tank closure approach would be to develop a scientific basis for tank closure which may allow greater waste volumes to be left in the tanks post-retrieval, while increasing protectiveness to human health and the environment. The approach is to characterize complex residual tank waste solids, measure contaminant release rates as a function of chemical environment, and build mechanistic release models.

### INTRODUCTION

Closure of the 230 remaining active single- and double-shell underground storage tanks poses unique technical, regulatory, and resource challenges at four sites across the DOE complex: the Savannah River Site (SRS), the Idaho National Laboratory (INL), West Valley Demonstration Project (WVDP) and Hanford. This is, in part, because of the different nature of the waste at the various sites, the composition and types of tanks, as well as the different regulatory regimes at each site. Despite the complexities, tank closure progress has been made as illustrated by Table I.

TABLE I. Tank Closure Status<sup>a</sup>

	<b>Hanford</b>	<b>SRS</b>	<b>Idaho</b>	<b>West Valley</b>
<b>Total Number of Tanks</b>	177	51	15	4
<b>Closure in Progress</b>	17	9	0	4
<b>Grouted and Stabilized</b>	0	6	11	0

At Hanford, active retrieval operations have been concluded with sampling and characterization completed for eleven tanks from the C-Tank Farm and one from the S-Tank Farm, for a total of twelve. Hanford is currently in active retrieval for three tanks in the C tank farm.

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<sup>a</sup> Data as of December 2014

At SRS, six out of fifty-one tanks have been closed and grouted. Furthermore, one tank has been isolated awaiting grouting while another is undergoing sampling and characterization. Four tanks have completed heel retrieval while an additional three tanks have had bulk retrieval finished. During mechanical bulk removal, typically over 95% of the radioactive inventory in the tank is removed. Subsequently, cleaning methods typically allow for the removal of 99% of the radioactive inventory.

At the WVDP, the HLW has been vitrified. A tank and vault drying system was installed in 2010 to evaporate liquids. Three of the four WVDP tanks are dry. Evaporation in the fourth tank, Tank 8D-4, has been slowed to allow for more time to resolve the disposition of the tank and its contents before the sludge is uncovered. Tank 8D-4 contains waste from the vitrification process, approximately 14,000 liters of liquid and 4,200 liters of sludge. The West Valley tank closure decision (and schedule) is tied to the West Valley Phase 2 NEPA decision. In its April 2010 Record of Decision (ROD), DOE committed to making a decision on Phase 2 of the project within 10 years (or by 2020) which will include the milestones for grouting the tanks.

At the INL, the site has 15 stainless steel tanks of which 11 are closed and grouted. The contents of the 11 tanks were emptied into one of the four remaining tanks. The tanks were then washed and sampled to confirm that closure standards were met. After confirmation sampling, the tanks, tank vaults and ancillary piping were grouted. Of the four remaining 1.1 million liter tanks, three tanks hold liquid sodium bearing waste and one tank is empty and held as a reserve. The waste will be treated at the Integrated Waste Treatment Unit (IWTU) once the facility is operational. After the four remaining tanks are emptied, they will be closed using the same process that was used on the previously closed 11 tanks.

## **APPROACHES AND TECHNOLOGIES FOR TANK WASTE CHARACTERIZATION, RETRIEVAL, AND TANK CLEANING**

Despite the progress, DOE has much yet to accomplish to close the Department's waste tanks. There is still a need to increase efficiencies and correspondingly to decrease the costs of tank closure. These challenges are often exacerbated by the necessity of fitting the tank cleaning apparatus through the tank risers.

### **Heel Retrieval – Mechanical Cleaning Approaches**

DOE commonly uses mechanical removal techniques that use liquids (e.g., acids or water) to loosen the heel thereby enabling retrieval. Spraying and lancing within the waste tanks is performed by inserting a nozzle through an open riser in the waste tank and directing the liquid at a targeted location. Lancing refers to a higher pressure, more concentrated spray pattern aimed at breaking-up or moving the solids within the waste tank.

DOE has adapted and successfully used a vacuum heel retrieval technology in the cleaning of unobstructed SRS Type IV tanks, Tanks 18 and 19. This technology used a cleaning device, called a Mantis, consisting of a mechanical crawler along with an ultra-high-pressure water eductor to vacuum residual solids and transports the slurry to a receipt tank.



Fig. 1. Mantis Device

Hanford has also adapted a FoldTrack device, originally used for cleaning sludges in oil tankers and conveyance equipment, for use inside of unobstructed tanks. Similar in concept to the Mantis, the FoldTrack mechanically breaks up chunks of waste, moving solids to the pump inlet. The apparatus can collapse, i.e. fold to fit through tank risers. The Foldtrack has nozzles to spray high-pressure water directly at the waste.



Fig. 2 FoldTrack

At Hanford, the Mobile Arm Retrieval System (MARS) supports the retrieval of radioactive and chemical waste from underground single shell storage tanks. The MARS was designed using a standardized platform thereby enabling multiple retrieval technologies. There are two distinct retrieval mechanisms, the MARS-Sluicing (MARS-S) and the MARS-Vacuum (MARS-V). The MARS-S routes pressurized fluids through spray nozzles to loosen waste materials. The MARS-V minimizes the amount of liquid in the tank by directing pressurized fluids through an eductor nozzle while drawing a vacuum on the waste material. The system can use recycled liquid waste in a closed loop. During testing, the MARS-V demonstrated the ability to remove sludge, small rocks, sand and the hard-packed waste found at the bottom of some tanks.

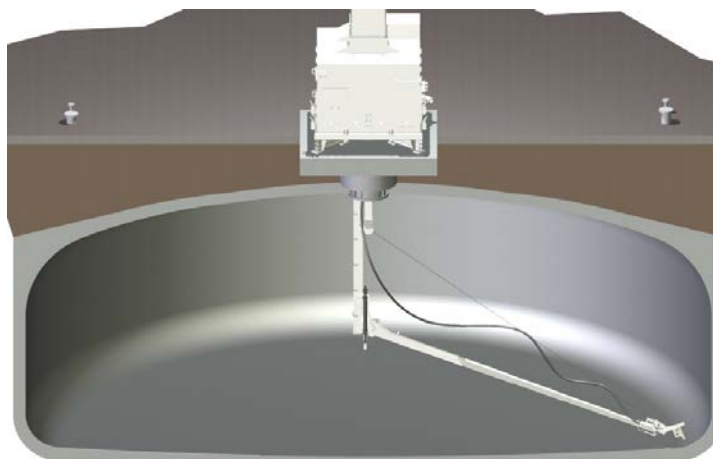


Fig. 3. Schematic of the Mobile Arm Retrieval System (MARS)

### Heel Retrieval – Chemical Cleaning Approaches

SRS has used two tank chemical cleaning technologies: Low Temperature Aluminum Dissolution (LTAD) and Bulk Oxalic Acid Cleaning (BOAC). Recent heel retrieval using chemical cleaning on SRS Tank 12 was very successful with regard to sludge heel (especially for Al, Fe, and U phases) and beta/gamma radionuclide removal. The chemical cleaning strategy used the following processing sequence: LTAD, washing, BOAC, and neutralization. Although chemical cleaning using these technologies has been shown to be effective, no disposition path has been identified for oxalate added during BOAC, and insoluble oxalate salts are accumulating within the SRS tank farm and waste processing facilities. Extensive sludge washing is required to remove moderately soluble sodium oxalate salts prior to sludge vitrification in the DWPF. In addition, the use of BOAC may result in tank residuals that potentially include moderately soluble oxalate salts with a different mobility within the environment than the original waste sludge phases. Consequently, oxalate additions to the tank farm need to be minimized by the use of supplementary acids to assist sludge removal or the use of other cleaning reagents or processing strategies.

Neither LTAD nor BOAC are highly effective at removing certain secondary metal components of HLW sludge; specifically, Hg, Ni, and Mn which are not effectively removed in the baseline chemical cleaning flow sheet. If removal of these secondary components is required for tank closure, then methods need to be developed to solubilize these constituents. It may be useful to develop methods for the removal of these constituents even if removal of these species is not generally required for tank closure, since specific tanks may have different cleaning requirements.

Because of the downstream challenges posed by BOAC, Hanford is moving away from this technique. Instead Hanford is using modified sluicing with tank supernate to mobilize and retrieve the residuals. The Hanford tank waste is more diverse than SRS waste due to the variety of different chemical processes that were used at Hanford. Retrieval of Hanford HLW tank heels may require alternative approaches.

Because of the difficulties and limitations inherent in BOAC, the Tailored Chemical Cleaning Project was initiated at the Savannah River National Laboratory (SRNL) to investigate alternatives to mechanical and chemical tank cleaning to remove residual heels. The project is focused on the optimization of existing tank cleaning methods as well as further development of promising methods identified in previous studies.

The removal of alpha emitting radionuclides present at low concentrations such as Pu, Am, and Np, can be problematic as these actinides are not highly soluble in currently utilized chemical cleaning reagents. Scoping studies have revealed promising methods to dissolve the actinides within the HLW tank heels. Oxidation of the actinides with permanganate in either strong caustic or dilute acidic solutions results in the dissolution of the oxy/hydroxide phases of these metals in the absence of major sludge phases. Either of these two permanganate-based methods for alpha removal might be suitable for incorporation into a chemical cleaning flow sheet, though they would likely be utilized at different times in the processing sequence. Use of permanganate-based methods results in the addition of manganese oxide solids to the waste and may affect downstream processing, so minimization of permanganate additions is necessary.

The goal of the Tailored Chemical Cleaning Project is to develop a strategy for the optimized retrieval of waste tank heels involving minimal oxalate additions and the retrieval of alpha-emitting radionuclides.

### **Waste Characterization**

Waste sampling and characterization is required to assess the chemical and radiological characteristics of the residual wastes and the fixed contamination in tanks. The results from characterization are used to develop a technical basis for decision making to ensure that closure actions are effective in protecting human health and the environment. Many of the radionuclide analyses involve multiple cycles of radiochemical separations to ensure removal of interfering nuclides and to achieve low minimum detection limits. In many cases, the time requirements for completion of the radionuclide analyses are several months, and the respective costs are commensurately high. Key challenges to characterize waste function include [1]:

*Equipment For Waste Characterization* – Waste retrieval sampling is generally costly. In-situ tools and equipment that could perform sampling characterization activities remotely, in real time, would be desirable because they lend themselves to quicker results and an ability to allow the characterization to direct early decision points for retrieval. There is also a challenge to having a suite of tools that reach and access difficult to locate areas of the tank or system whether for in-situ or laboratory analysis.

*Slow Turnaround of Sample Analyses* – Tank waste characterization significantly increases the time required for tank closure. Faster turnaround of laboratory analyses is challenged by limited resources and lengthy analytical durations. Challenges exist in methods, equipment and expertise needed for waste tank characterization that ultimately impact retrieval and closure campaigns.

*Inadequate Detection Limits for Species of Interest* – Often the concentration of species needs to be measured at a much lower level than the capability of the instruments available. In some cases, the insufficient detection limit is due to the limits of the equipment. At other times, it is due to the high radiological dose of the sample, which requires dilution prior to analysis. Characterization tools need to be developed to achieve lower detection limits for these species of interest.

To address these issues, the Cost Effective Tank Waste Characterization project was initiated to develop strategies and technologies to optimize tank waste characterization. The goal is to implement programmatic changes that accelerate tank waste processing and tank closure schedules, while at the same time reduce characterization costs. The specific objectives are: a) to gain a sound understanding of the relative costs, time requirements, and relevancy of current characterization activities/practices; b) to assess potential alternative characterization methodologies; and c) to identify opportunities for improving characterization practices in the context of reducing cost and schedule. The initial efforts will rely heavily on baseline information drawn from experience characterizing SRS HLW sludge, salt, and tank closure residue samples and will:

- Identify characterization activities driving cost and schedule;
- Investigate streamlining of characterization requirements based on the relative constituent risks (reduce characterization requirements for "low risk" constituents );
- Determine the relative usefulness of laboratory analyses, waste receipt history, process knowledge, scaling factors, and other potential characterization bases;
- Use differences between sludge, salt, and post-cleaning residue to hone characterization needs as a function of waste type; and
- Investigate alternative characterization methods holding promise for being less costly and/or less time consuming.

These activities may lead to more cost effective and practical tank waste characterization programs. Although it is desirable to minimize the number of analyses and to streamline the characterization process, it is also necessary to conduct a sufficient chemical characterization of the heel to support the Performance Assessment (PA) results.

## **ALTERNATIVE TANK CLOSURE OPPORTUNITIES**

Current tank waste retrieval requirements are based on volume or the more subjective criteria of as much waste as reasonably feasible. Because these retrieval requirements are not directly correlated with the risk associated with the residual tank waste, an opportunity exists to consider alternative tank closure strategies that are informed by risk rather than subjective criteria.

Due to the recalcitrant nature of tank waste solids, a typical tank waste retrieval campaign at both Hanford and SRS may cost in excess of \$20 million per tank. The current volume-based

retrieval goals at Hanford were developed independent of the risk posed by the residual waste left in the Single Shell Tanks (SSTs) post-retrieval, and as a result, the State has agreed to an alternative of using up to three retrieval technologies deployed in a serial fashion to demonstrate that DOE removed the residuals to the extent practical.

Because tank retrieval and closure will require sustained investments of public resources for many decades, DOE needs to collect information that will ensure that those public resources are applied in an efficient and protective manner. Hanford has initiated a Closure Demonstration Project to assist in achieving the closure milestone by working through technical and regulatory issues with regulatory agencies.

These INL, SRS and Hanford retrievals, residuals sampling and analysis, and closure actions provide an opportunity for improving our understanding of the risk and cost implications of retrieval and closure methods that can be applied to subsequent tank farm closures. Gaps in the technical foundation and modelling supporting the tank closure decision processes have traditionally resulted in a number of overly conservative assumptions in tank closure PA's. For example, a lack of a full understanding of the physical and chemical processes that control radionuclide release from residual waste has led to the use of simplified source term release models which result in highly conservative estimates and result in conservatism in decision making regarding the degree of cleanliness necessary prior to tank closure. Recent experimental results and new models on concrete performance and groundwater movement are improving the assumptions used in PA's and decreasing the uncertainties, allowing for a better understanding of the expected circumstances. In addition, new capabilities being developed as part of the Advanced Simulation Capability for Environmental Management (ASCEM) Project are well suited for reducing the need for conservative assumptions in PA's. These capabilities are currently being deployed to provide technical underpinnings for both the Hanford Waste Management Area C and the SRS H-Tank farm PA's.

Recent work has demonstrated that development of scientifically based mechanistic release models is critical to understanding contaminant release from post-retrieved tank waste residuals [2]. For example, studies of uranium release from Hanford's C-200 series tanks have shown that after the retrieval goal was achieved, uranium release from the residual waste is expected to be over 3 orders of magnitude above the Maximum Contaminant Limit (MCL). With the aid of a thermodynamic solubility release model developed to predict future uranium release from tank residuals, a method was designed to render uranium insoluble within the residual waste through the addition of a small layer of lime (CaO) on top of the tank residuals prior to closure [3].

Implementing a risk informed decision making approach to tank closure requires an integrated laboratory and modelling program to develop a strong technical foundation for retrieval and closure. Requirements of a successful program include:

- Quantifying the long-term risk reduction benefits of varying degrees of tank retrieval and the differential effect of alternative retrieval methods and end points (e.g., sluicing vs. chemical dissolution versus. dry mechanical retrieval methods, etc.);

- Accounting for the physical and chemical processes controlling radionuclide release from the source term resulting from the use of different technologies in retrieval steps; and
- Determining the retrieval endpoints on the basis of risk associated with the fate and transport of radionuclides through the vadose zone to the point of compliance.

## CONCLUSIONS

Closure of the underground storage tanks at sites across the DOE complex poses unique technical, regulatory, and resource challenges. Sludge waste heels invariably remain in the tanks after the bulk of the waste has been retrieved, and reducing the volume of these heels becomes increasingly difficult as their volumes decline. There is no one single best process for tank closure applicable to all the sites or even within a site. The tank closure approach has to be determined on a case-by-case basis. The optimal retrieval process used for a particular tank will be dependent upon the chemistry and physical characteristics of the tank waste.

The ongoing chemical cleaning project will provide a strategy for optimized retrieval of SRS waste tank heels involving minimal oxalate additions and the retrieval of alpha-emitting radionuclides. In addition, new chemical cleaning approaches for the retrieval of more diverse Hanford wastes will need to be identified and tested. Similarly, the ongoing tank waste characterization efforts may lead to more cost effective and practical tank waste characterization programs.

Quantitative, scientifically-defensible and laboratory parameterized models of contaminant release from tank residuals are needed to inform risk informed tank closure decisions. For example, quantifying the long-term risk reduction benefits as a function of the amount of remaining tank heel may provide an alternative to volume or limits of technology based tank closure metrics currently being used at SRS and Hanford. Furthermore, the chemical stabilization of residual tank waste combined with risk informed closure could support retrieval endpoints other than those based on volume or the “limits of technology” and may result in a greater level of protection to human health and the environment.

Near term activities will involve establishing the sample characterization protocols needed to develop contaminant release models for residual tank waste samples, identifying interim stabilization approaches for residual tank waste solids, and quantifying the total risk reduction and cost savings that could be realized through the implementation of a risk informed retrieval and closure strategy.



## REFERENCES

1. Fellingner, A.P. et al., EM-21 Retrieval Knowledge Center: Waste Retrieval Challenges, SRNL-STI-2009-00231, April 2009.
2. Cantrell KJ, KC Carrol, EC Buck, D Neiner and KN Geiszler. 2013. Single-Pass Flow-Through Test Elucidation of Weathering Behavior and Evaluation of Contaminant Release Models for Hanford Tank Residual Radioactive Waste. *Applied Geochem.* 28:119-127.
3. Cantrell KJ, WJ Deutsch, and MJ Lindberg. 2011. Thermodynamic Model for Uranium Release from Hanford Tank Residual Waste. *Environ. Sci. Tech.* 45(4):1473-1480.