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Revision 1

Waste Removal Technology Baseline: Technology Development Description

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15 June 2011

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ACRONYMS/ABREVIATIONS

ADMP	Advanced Design Mixer Pump
AHP	Analytical Hierarchy Process
BWR	Bulk Waste Removal
DOE	United States Department of Energy
DWPF	Defense Waste Processing Facility
ECC	Enhanced Chemical Cleaning
ECR	Effective Cleaning Radius
EDTA	Ethylenediaminetetraacetic Acid
EMMA	Easily Manipulated Mechanical Arm
FTF	F-Tank Farm
HLW	High-Level Waste
HTF	H-Tank Farm
HM	H-Modified
HRR	Highly Radioactive Radionuclide
MFB	Mechanical Feed-and-Bleed
OA	Oxalic Acid
PUREX	Plutonium Uranium Extraction
RKC	Retrieval Knowledge Center
SEE	Systems Engineering Evaluation
SDF	Saltstone Disposal Facility
SLP	Standard Slurry Pump
SMP	Submersible Mixer Pump
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
TFA	Tanks Focus Area
THOREX	Thorium Extraction
WM	Waste Management

Note: The document's units of measure are common to the technical discipline target audience and have not been defined in the above list.

1.0 INTRODUCTION

The development of the tools and techniques used to clean out high-level waste (HLW) tanks at the Savannah River Site (SRS) evolved over decades to the methods now employed as the baseline technologies. The technologies represent knowledge gained through research and experience that began in the mid-1960s and continues through today. The waste collected in the waste tanks at SRS represents almost 60 years of nuclear materials reprocessing.

This report summarizes the baseline technologies used to remove waste from the waste tanks with emphasis placed on removing sludge from waste tanks. The evolution and technical maturation selection of these technologies is briefly described, which includes a historical perspective. The report also discusses evaluating and advancing new technologies when identified. The actual process used for waste removal can vary depending on the service history of the waste tank system, the physical characteristics of the waste remaining, the physical configuration of the waste tank system, and the timing of the waste removal actions.

2.0 DESCRIPTION OF THE SAVANNAH RIVER SITE PROCESS

The various nuclear processes used at SRS had a direct bearing on the radiochemical and mechanical properties of the waste in each waste tank. The physical behavior of the waste differs slightly from waste tank to waste tank. This section describes the liquid waste process, the waste tanks, the type of waste encountered in the waste tanks and the physical characteristics of the waste forms.

2.1 Location and Mission

SRS is a 300-square mile industrial facility constructed in the early 1950s. The SRS is located in western South Carolina and covers portions of Aiken, Barnwell, and Allendale counties. The SRS produced weapons-grade plutonium, uranium, and tritium as part of the United States Department of Energy (DOE) national defense mission. Environmental remediation at SRS includes cleaning and closing nuclear waste tanks.

2.2 Liquid Waste Process Summary

Radioactive waste at SRS is generated from the chemical separations facilities and is present in the tank farms as insoluble solid compounds and water-soluble salts. Since the first waste receipt in Tank 1 in 1954, SRS has generated over 150 million gallons of high level nuclear waste. Evaporation operations reduced this volume to the present inventory of about 38 million gallons.

The waste is stored in 49 underground waste tanks in F-Tank Farm (FTF) and H-Tank Farm (HTF). Two of the original 51 waste tanks were operationally closed in 1997. While stored in the waste tanks, the insoluble solids settle and accumulate on the bottom in the form of sludge. The liquid volume is reduced by evaporating excess water. The concentrated salts crystallize forming hard, but porous, saltcake. Tank farm facilities also pre-treat the accumulated sludge and salt solutions to facilitate further processing at other SRS treatment facilities (i.e., Defense Waste Processing Facility (DWPF) and Saltstone Disposal Facility (SDF)). These treatment facilities convert the sludge and supernatant liquid to more stable forms suitable for permanent disposal. To date, removing salt waste from a waste tank has not posed the technological hurdle or difficulty as removing sludge. Typically, saltcake is removed by dissolving with fresh water and then transferring the solution to another waste tank. In some cases, agitating the waste tank with mixing pumps expedites the dissolution process. Salt dissolution leaves insoluble solids and sludges in the waste tank, which is then managed as a sludge removal process. For this reason, this report focuses on the baseline technologies and selection of sludge and solids removal.

2.3 Separations Processes and the Effects on Sludge Properties

The types of nuclear reprocessing carried out at SRS include plutonium uranium extraction (PUREX), H-Modified (HM), and thorium extraction (THOREX). The waste products from these processes differ slightly in composition, but each is a result of a liquid-liquid organic extraction process with ion exchange. PUREX removes plutonium, uranium, and technetium isotopes from irradiated fuel rods (dissolved in concentrated nitric acid). Similar to PUREX, HM is slightly modified to process limited amounts of other isotopes such as neptunium and californium. Specialized campaigns to recover americium and curium were also performed. THOREX is a method to remove thorium isotopes. Each process carried out in the SRS separations facilities or canyons, is acidic. Because the waste tanks are made of carbon steel, the waste stream is neutralized with sodium hydroxide and corrosion inhibited with sodium nitrite in the separation facilities before being sent to the tank farms. The neutralization reaction creates the salts and precipitates the solids.

The settled solids form sludge and follows Bingham plastic rheology model, a fluid that possesses a yield stress. In other words, a minimum amount of force has to be applied before the fluid starts to move (i.e., toothpaste in a tube). Figure 2.3-1 shows the differences between Bingham plastic behavior and other fluids.

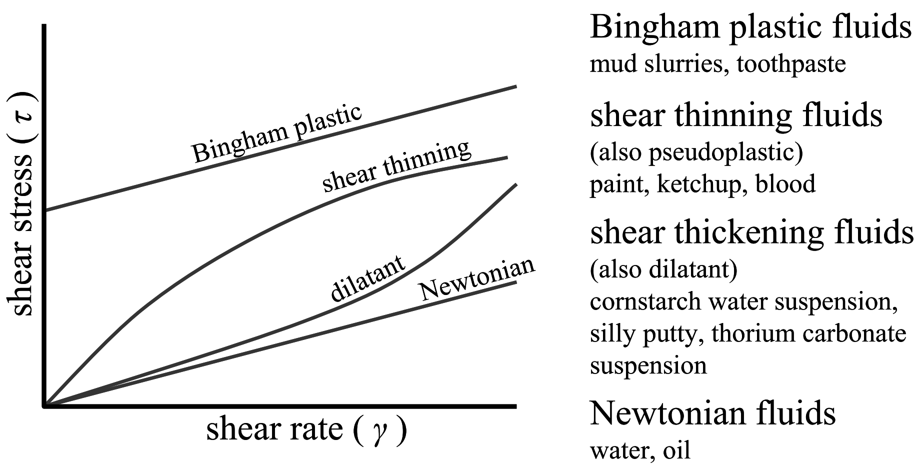

Figure 2.3-1. Stress vs. rate relationship for various fluids

The composition of the sludge solids depends primarily on the original separations process. For example, the solids waste products from the PUREX process are predominately iron-based compounds with small amounts of depleted uranium and trace amounts of plutonium. The PUREX sludge is characteristically dark brown with quick-settling solid particles as illustrated in Figure 2.3-2. The HM process conversely produces less dense sludge that is aluminum-based, with slight iron, with a longer settle time. Settled HM sludge is difficult to re-suspend because of the cohesiveness of the small particles.

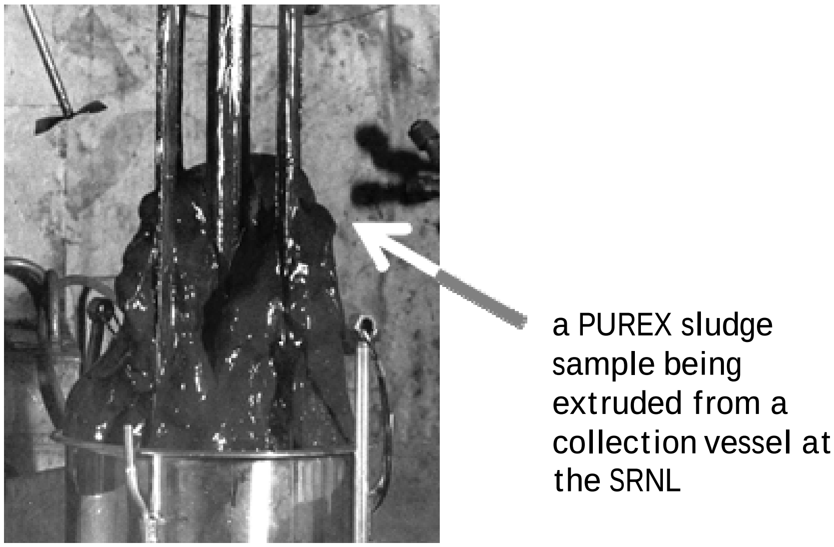

Figure 2.3-2. PUREX sludge sample

Waste solids from the THOREX process exhibit unusually high yield stresses at relatively low concentrations. At high concentrations, THOREX sludge behaves like a gel and must be diluted several fold to make the slurry pumpable.[1] There is a small amount of this type of sludge in several of the HTF waste tanks.[2]

2.4 Waste Tank Description

The waste tanks are comprised of four types. The Type I tanks are the oldest with a nominal capacity of 750,000 gallons and a 5-foot high secondary containment steel liner within a concrete vault. The Type I tanks are approximately 75 feet in diameter. The next storage class is the Type II tanks with a storage capacity of 1,030,000 gallons with an 85-foot diameter. The Type II tanks also have a 5-foot high secondary containment steel liner within a concrete vault. The most modern waste tanks are the 1,300,000-gallon capacity Type III/IIIA tanks. They boast a full secondary containment, an integral cooling and ventilation system, heat-treated carbon steel liners and numerous access openings. The total length of cooling pipes in a waste tank is approximately four miles. Each Type I, II, and III waste tank has an intertwining array of 2-inch diameter cooling pipes. These pipes interfere with waste removal and waste tank closure activities. The 1,300,000-gallon un-cooled waste tanks make up the fourth design (often referred to as the Type IV tanks). The un-cooled waste tank variety has a self-supporting dome roof and no internal cooling coils. The un-cooled waste tanks were used primarily to store low activity waste. All the waste tanks are either subterranean or surrounded and covered with soil for radiation shielding.

[1] C. B. Goodlett, "Chemical Processing of Irradiated Thorium Handling of First Cycle Aqueous Wastes – Supplemental Data." Letter to E. B. Sheldon (DPST-64-238), pp. 2-3.
[2] H. Q. Tran, *Tank Radioactive and Non-radioactive Inventories (8/14/2007)*, (LWO-LWE-2007-00250), p. 54.

2.5 Sludge Behavior in Various Waste Tanks

As discussed in Section 2.3, SRS engaged in several radioactive separations processes, each producing waste streams with different physical properties. This resulted in waste tanks containing waste that had drastically different fluid behavior. Figure 2.5-1 illustrates how the yield stress can differ in solids concentration from one waste tank to another.

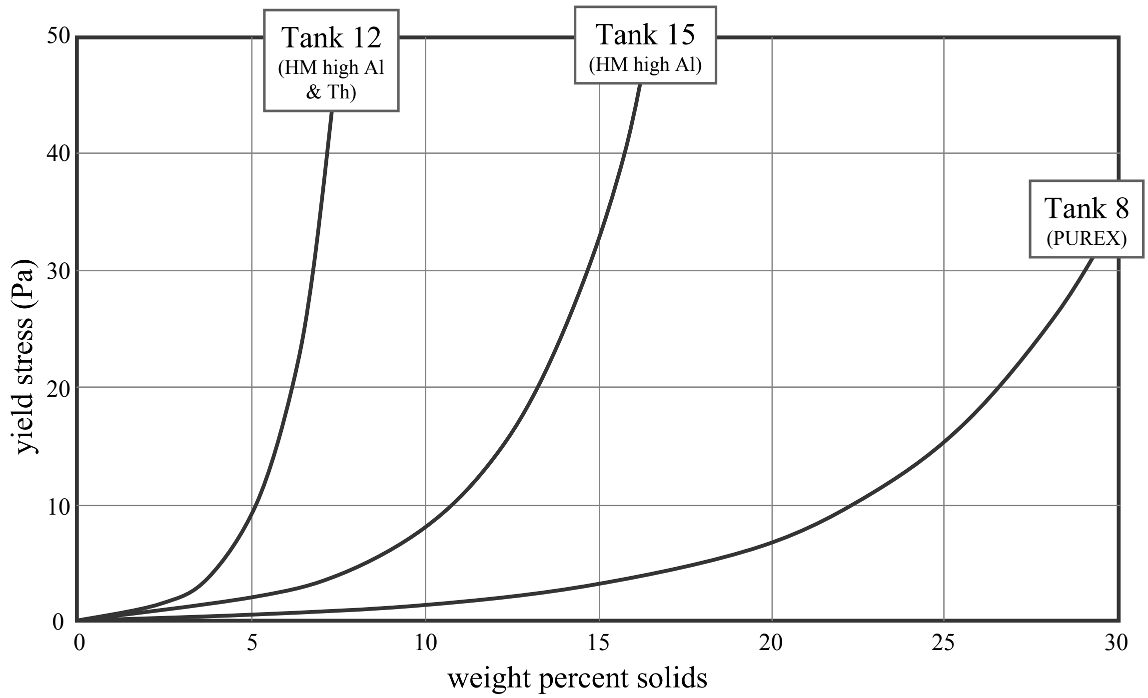

Figure 2.5-1. Yield stress vs. solids concentration for various waste tanks

The varying sludge behavior combined with the flat bottom, numerous interior interferences, and limited numbers of waste tank top access openings hinder waste removal efforts. The waste radiotoxicity precludes direct contact and promotes remote handling methods. The technologies explored by SRS workers involved variations of mixing, chemical dissolution, mechanical removal and other remote robotic techniques.

3.0 Technology Selection Process

In the waste removal technology selection process, a structured approach (e.g., Systems Engineering Evaluation (SEE)) identifies and compares viable alternatives that meet the defined functions and requirements for waste removal in the specific application. The SEE is an example of a technology selection process successfully used at SRS.[3] A SEE is a formal analysis and is based on a set of weighted decision criteria working within a group of constraints. A sensitivity analysis can assist in the proper selection of a preferred alternative.

A typical technology selection method will include some or all of the following elements:

- Identify a group of subject matter experts, informed individuals, customers, and stakeholders to identify and assess viable waste removal options.
- Identify functions to be met, the project requirements, and the constraints. A criterion for a technology includes a reasonable likelihood of achieving estimated removal results.
- Identify alternatives that can perform the functions.
- Determine the viability of the alternatives to satisfy requirements.
- Establish the weight (or importance) of the criteria to evaluate alternatives.
- Evaluate the alternatives against the criteria.
- Select a preferred alternative (or group of alternatives).

This section describes how a technology is evaluated, introduced, and used as a baseline method for waste removal. This includes establishing a set of physical and operational constraints, deciding on selection criteria based on those constraints and performance goals, identifying new technologies from a number of sources and evaluating those technologies against the criteria using proven decision-making methods.

3.1 Program Requirements

This section describes the waste removal and tank closure program requirements. The requirements must consider regulatory, facility, and fiscal parameters.

3.1.1 Regulatory Requirements

In order to proceed with closure of the SRS tank farms, DOE Manual 435.1-1 and DOE Guide 435.1-1 requires the issuance of Tier 1 Authorization defining the parameters, approach, plans, and requirements by which tank closure activities will be accomplished. The DOE approval to proceed with permanent stabilization of a specific waste tank will be based on Tier 2 Closure Authorization which documents that all the criteria required by the Tier 1 Closure Authorization have been met. Documentation to support Tier 2 Closure Authorization includes, among other things, demonstration of conformance with the applicable Determination by the Secretary of Energy, in consultation with the NRC, under Section 3116(a) of the Ronald W. Reagan National Defense Authorization Act for Fiscal Year 2005 (NDAA) and the supporting Basis Document.

The SRS Federal Facilities Agreement (FFA) establishes that, among other things, the SRS waste tanks that do not meet secondary containment standards (Type I and IV) must be removed from service according to the FFA schedule.

[3] M. J. Cercy, *Systems Engineering Methodology Guidance Manual* (WSRC-IM-98-00033, Rev 14), Appendix A.

Tank waste storage and removal operations are governed by an SCDHEC industrial wastewater construction permit. Removal from service and stabilization of the waste tanks will be carried out pursuant to a State-approved closure plan. The closure plan for FTF, *Industrial Wastewater General Closure Plan for F-Area Waste Tank Systems*[4], has been issued, a similar document for HTF will be developed in the future.

The requirements for all of the above listed programs must be considered as part of the technology selection process

3.1.2 Fiscal and Schedule Constraints

Depending on the individual project scope, waste removal processes and equipment come under the purview of DOE Order 430.1B, Change 1, *Real Property Asset Management*. Funding for waste removal activities is contingent on the federal budget with the priority ranked among other DOE environmental and defense programs. Therefore, waste removal methods must be chosen with conscientious stewardship and the selection of those methods must include a cost benefits analysis. In addition, the schedule for deploying methods depends on regulatory and contractual aspects. Some of the equipment has long procurement and acquisition times (e.g., exceeding two years).

3.1.3 Tank Storage Space

The tank farms have a limited amount of storage space. Performing a waste removal program requires more tank space than the waste being removed. That is because additional dilution and mixing liquid is used to aid in the mixing evolutions. Detailed system integration planning is completed to coordinate the use of waste tank space meeting processing and regulatory commitments. The use of excessive volumes of liquid is prohibited. Many industrial tank-cleaning processes use large volumes of liquid or solvents. Space limitations reduce the possibility of adapting commercial tank cleaning methods. The majority of SRS working waste tanks are at or near full capacity. Figure 3.1-1 illustrates Tank 41 filled to capacity with saltcake.

[4] Birk, M. B., *Industrial Wastewater General Closure Plan for F-Area Waste Tank Systems*, (LWO-RIP-2009-00009), 2011.

Figure 3.1-1. Tank 41 at full capacity (2003)

3.1.4 Downstream Effects

The process streams resulting from waste removal operations must be compatible with the existing facility, and congruent with the feed streams of DWPF, Salt Waste Processing Facility, and the SDF. In other words, the streams must have a viable disposal path.

3.1.5 Job Waste Disposal

Similar to downstream effects, the equipment used for waste removal must also have a disposal path. Exacerbating this limitation is the equipment is often large due to the vastness of the waste tanks (e.g., the mixer pumps are 45 feet long). This presents a problem for processing and disposal. Accordingly, waste removal technologies must consider long-term equipment disposition in the design.

3.1.6 Waste Tank Integrity

The earliest waste tanks are approximately 60 years old. The age of the waste tank requires consideration as removal methods are selected. Since the waste tanks are constructed of carbon steel, the use of strong acids or oxidizers to dissolve waste residue is precluded. Many of the older waste tanks (Type I and II tanks) have leaked from the primary vessel into the secondary pan through cracks caused by stress corrosion cracking. Although the conditions that caused the cracks have been abated, the cracks in these waste tanks are still present. Waste removal efforts often involve disturbing former leaks sites, while revealing leak sites not previously identified, therefore, compensatory actions are put in place during waste removal activities, ensuring waste is contained and to avoid the spread of contamination. The newer style waste tanks (Type III/IIIA) have no recorded leaks.

3.1.7 Natural Phenomena

The tank farms are located outside without the protection of a containment structure. Equipment must be able to withstand high summer temperatures prevalent in the Southern United States as well as subfreezing conditions and hurricane force winds and rain.

Containment structures in the tank farm must be designed to withstand all types of natural phenomena hazards.

3.1.8 Transfer System Integrity

The pipeline network connecting the waste tanks is aged. Most of the pipes were designed and constructed without consideration for the high pressures that are probable when transporting high yield stress slurries. As a result, viable waste removal technologies must consider the limitations of the piping used for transporting the waste.

3.2 Constraints

This section describes the physical constraints to which the waste removal and tank closure program must adhere.

3.2.1 Waste Tank Interior Inferences

Waste removal methods must be able to work around (or with) interferences such as interior support columns, horizontal, and vertical runs of cooling coils, discarded equipment and installed waste tank monitoring equipment. Refer to Figure 3.2-1 for a depiction of congestion formed by the array of cooling coils in a typical Type I tank.

Figure 3.2-1. Tank 4 cooling coils (1953 Pre-Operation)

3.2.2 Remote and Limited Tank Interior Access

The waste tank bottom is 45 to 50 feet below ground level. There is limited access to the tank interior as illustrated in Figure 3.2-2. The majority of the risers are less than 2 feet in diameter. For the Type I tanks, the risers can be as long as 10 feet. Construction tolerances resulted in waste tank risers not being uniform in diameter (eccentric). Waste removal methods must be sufficiently tolerant to address limited waste tank accessibility and to account for field discrepancies in waste tank dimensions. Furthermore, installing additional risers on the Type I and II Tanks, although feasible, would be a difficult endeavor because of the high radiation fields and remote access to the tank tops. However, there are advantages to having more tank openings, and SRS is continuing to investigate cost-effective and safe methods of drilling new openings through the tops of these waste tanks.

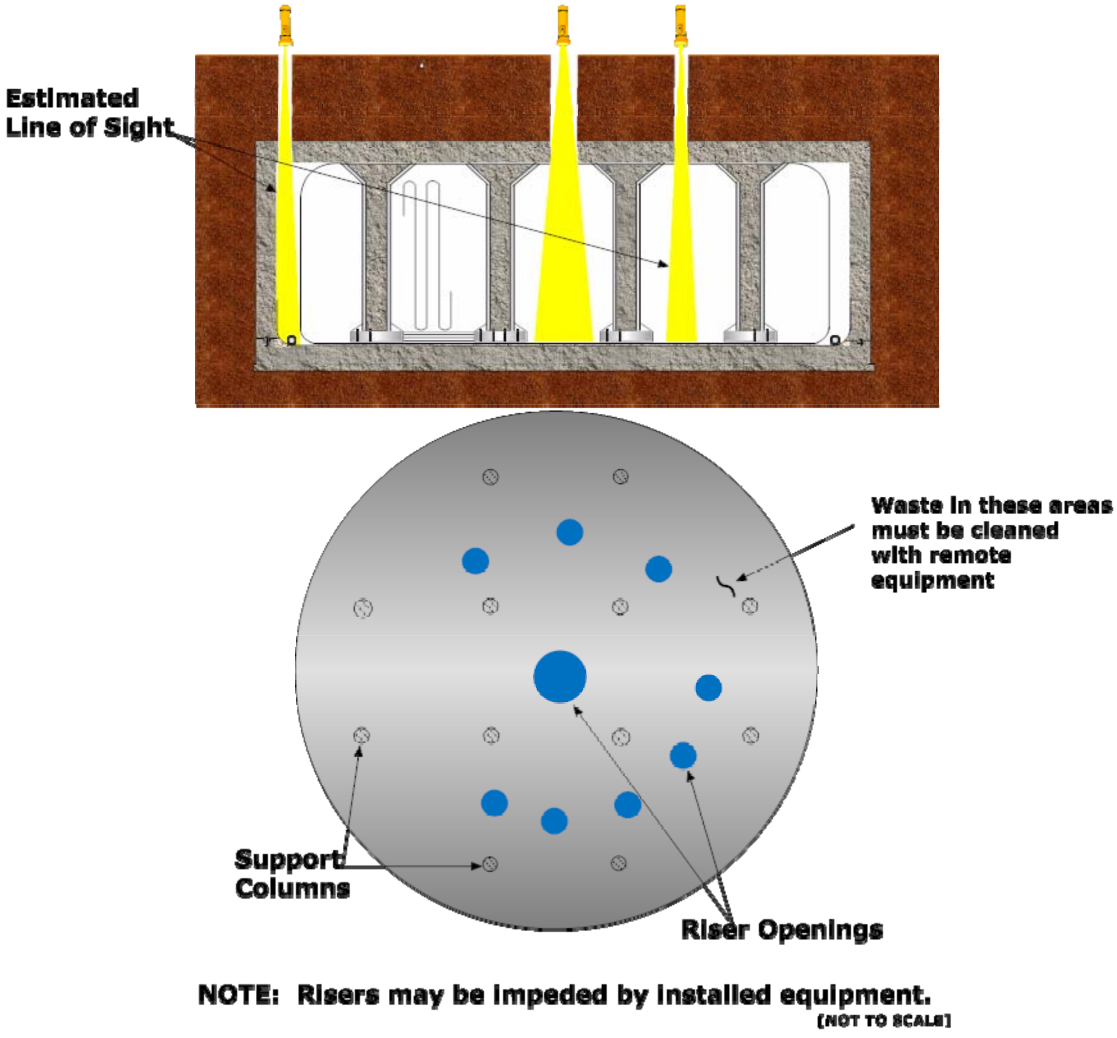

Figure 3.2-2. Access area for a typical Type I waste tank

For Type IV tanks 18 and 19, new risers were installed to support waste removal activities.[5] The Type IV tanks, compared to Type I and II tanks, have easier access to the tank top and the roof thickness is less. An analysis of the cost versus benefits of installing a new riser will need to be evaluated for each tank.

3.2.3 Limited Tank Storage Space

Available waste storage space is limited in the tank farms. The majority of SRS working waste tanks are at or near full capacity.

3.2.4 Operational Conflicts

Waste tanks that are targeted for cleanup and closure reside in an operational facility. The tank farms (including waste tanks actively in a closure status) retain an operational posture that supports national defense programs. Equipment (e.g., cranes), facilities (e.g., evaporation

[5] M. B. Birk, *Documentation of Removal of Highly Radioactive Radionuclides in Waste Tanks 18 and 19*, (SRR-CWDA-2011-00091), 2011.

systems), and resources (e.g., radiation technicians) are often shared. Consequently, waste removal technologies must consider the impacts to other ongoing operations.

3.3 Hazards

This section describes the unique hazards workers must endure when performing waste removal and tank closure operations.

3.3.1 Radiological Contamination

Radiation exposure and contamination potential require strict radiological control protocols surrounding work areas in close proximity to any waste tank opening. This requires that personnel and equipment be shielded from radiation fields and protected from the spread of contamination. Furthermore, equipment used for waste removal must have sufficient integrity to contain any radioactive material handled.

3.3.2 Radiation

Waste tank risers must be opened for any equipment installation. The radiation fields near an open riser are hazardous. Personnel must be protected using "as low as reasonably achievable" exposure principles. In addition, the ionizing radiation fields inside a waste tank are strong enough to damage certain types of equipment. Some commercial-grade elastomers and most electronic equipment have limited operating life when used inside a waste tank. Electrical motors, gaskets, seals, hoses and electronics must be specially designed to be radiation hardened (i.e., resilient in high radiation fields).

3.4 Systems Engineering Evaluations for a Technology Selection

Selection efforts through the years have employed different techniques and methods. The most commonly used are systems engineering tools. The term systems engineering can be traced to the Bell Telephone Laboratories in the early 1900s with major applications during World War II. The need to identify and manipulate the properties of a system as a whole, which in complex engineering projects may differ greatly from the sum of the parts' properties, motivated the Department of Defense, National Aeronautics, and Space Administration, and other industries to apply the discipline of a systems engineering process. The Apollo space program is a leading example of a systems engineering program. Option selection is one of the methods used in the systems engineering discipline. A tool used extensively for option selection is the analytical hierarchy process (AHP), which is described below in greater detail.

When a clear alternative is not available or when several alternatives must be considered, a more complex method of selection is required. Since 1998, SRS teams have been following the guidelines outlined in the *Systems Engineering Methodology Guidance Manual* (WSRC-IM-98-00033) for managing complex engineering decisions. Appendix A of the manual provides guidance for alternative studies, often referred to as a SEE. A SEE selects an alternative from two or more options constrained by specific functions, selection criteria, and requirements. After identifying the functions, requirements, and selection criteria, options are screened and then evaluated. One method of evaluation is the structured method of handling complex decisions called AHP. The AHP helps decision makers find the option that best suits the needs and understanding of the problem. The team first decomposes the problem into a hierarchy of attributes, each analyzed independently. The hierarchy elements can relate to any problem attribute that applies to the decision at hand. For waste removal, the team may select the attributes of safety, development schedule, deployment/operations schedule, development cost, technical/operational effectiveness, and technical/operational complexity.

With the hierarchy of attributes identified, the team systematically evaluates the options by comparing one to another, two at a time (called a pair wise comparison). The team uses concrete data, experience, and judgment to make the comparisons. During the comparisons, the preference of each option is compared to each of the other options for the selected evaluation criteria using values of importance on a scale from 1 to 10, with 10 having the highest importance.

In the final step of the process, the options numerical scores are calculated by considering each attribute's weight of importance. The score represents the option's relative ability to achieve the decision goal, and allows a straightforward and documented decision over the various available courses of action.

Generally the preferred option is clear if that option's score exceeds the others by 10% or greater. If not, then a sensitivity analysis is performed. The purpose of a sensitivity analysis is to validate the option evaluation and ranking of options that result from the decision process by demonstrating that small changes do not change the ranking. These small changes could occur for the option scores against the criterion weights. The sensitivity analysis evaluates the impacts of adjusting criterion weights up and down by approximately 10%. If these small changes do not affect the overall results then the analysis is insensitive to the alternative scores.

The following is a list of some examples of SEEs that used the AHP for evaluating technologies to perform waste removal:

- Tank 19 Waste Removal (1998)[6]
- Tank 18 Waste Removal (2001)[7]
- Waste Removal Balance of Program (2003)[8]
- Tanks 5 and 6 Heel Removal (2009)[9]

The SRS continues to evaluate new technologies for potential use in waste removal applications as they are developed. In addition, they will also evaluate, case-by-case, any special conditions that may occur during waste removal activities that may require application of additional or alternative technologies.

When a new method or approach is discovered, subject matter experts are assembled to review the new technology. A preliminary recommendation for a path forward is made. The path forward is usually one of the following recommendations:

- Do not pursue because of non-applicability or poor technical fit with the tank farm system.
- Pursue for further development and maturation. This indicates the new technology has promise for improving the highly radioactive radionuclide (HRR) removal performance of the baseline methods, but requires further study and refinement before being considered an option as part of the selection process.

[6] N. R. Davis, *Tank 19 Heel Removal Systems Engineering Plan*, (PIT-MISC-0040).
[7] G. E. Abell, *HLW Tank 18 Waste Removal Systems Engineering Evaluation*, (WSRC-RP-2001-00024).
[8] D. A. Zupon, *Waste Removal Balance of Program Systems Engineering Evaluation Report*, (G-ESR-G-00051).
[9] G. B. Clendenen (2009), *Strategy for Tanks 5 and 6 Phase II Mechanical Sludge Removal*, (SRR-CES-2009-00022).

- Adapt immediately into the baseline without significant maturation. This implies the technology is mature enough to integrate into the baseline without significant programmatic risk.

The decision to augment or replace a baseline technology for a waste removal process step (e.g., bulk waste removal (BWR), heel removal, etc.) is documented. The documentation assesses, at a minimum, the technologies' expected HRR removal capability, likelihood to meet the desired results effectively, costs, technical maturity, technical complexity, and reusability. Furthermore, some examples of costs considered are dose to workers, dose to public, financial costs, system-wide impacts (e.g., effects on downstream systems, generation of secondary waste streams), impacts to DOE's mission and schedule, transportation risks and radiological control requirements. If the Contract Performance Baseline is impacted by a change of a baseline technology, then the change must also be reviewed with DOE.

3.5 Identifying a New Technology

The source for new technologies is many-fold. Researchers have several information avenues at their disposal. Conferences, technical exchanges, and corporate sharing are ways to gather and share information.

3.5.1 Hanford-SRS-Idaho Waste Retrieval Technology Exchange

The Hanford-SRS-Idaho Waste Retrieval Technology Exchange is usually an annual symposium that began in 1998. Scientists, engineers, and managers present a new technology relating to HLW retrieval and management. The conference location alternates between cities on the west coast and east coast to encourage participation from each DOE complex site.

3.5.2 Waste Management Conference

The first Waste Management (WM) Conference was held in Tucson, Arizona in 1974. Government and private organizations undertake the global task of radioactive waste management at the yearly conference. The SRS conference participation extends back to the mid-1980s.

3.5.3 Special Technical Exchanges and Workshops

Periodically, special technical exchanges occur to address a current issue. Examples include:

- Tank Cleaning Technical Exchange (2006)
- Cementitious Material Workshop (2006)
- Aluminum Leaching Workshops (2006 and 2007)
- Slurry Retrieval Workshop (2008)
- Sellafield-SRS Workshop (2009)

These workshops help expand the body of knowledge regarding the science of waste removal and waste tank remediation. Participants at these exchanges often include some or all of the following representatives depending on the exchange: DOE, the national laboratories, contractors, vendors, regulatory agencies, and consultants. The Tank 18 and 19 Mantis used for mechanical heel removal was discovered at the Tank Cleaning Technical Exchange in

2006.[10] The exchanges provide a unified platform enabling engineers and researchers to develop a relationship for communication during project development.

3.5.4 Retrieval Knowledge Center

The Retrieval Knowledge Center (RKC) is a virtual information database related to the remediation of radioactive wastes from underground waste tanks throughout the DOE complex. The RKC goal is to provide user access to an electronic display (with bibliographic information) and document download capabilities, where available. The website for this information is www.rkc.pnl.gov. Presently, the RKC program is unfunded and new information is not being inserted, however, the database is still accessible.

3.5.5 United States Department of Energy Complex Site Communications

Employees from SRS, the Hanford Site, and Idaho National Laboratory participate in a bi-weekly conference call where information on technologies is shared and projects updated. Frequently, new contacts are established thereby shortening the virtual distance between these sites.

3.5.6 Corporate Knowledge

The companies involved with operation, closure, and remediation of the SRS tank farms have corporate experience in cleaning and closure in both nuclear and non-nuclear site decommissioning. There are several major contractors that are involved in remediation of industrial sites worldwide. These include URS Corporation, CH2M-Hill, Bechtel, and B&W.

3.6 Annual Review

As required by the *Industrial Wastewater General Closure Plan for F-Area Waste Tank Systems,*[11] DOE provides annual updates on new waste removal and characterization technologies to the South Carolina Department of Health and Environmental Control in a dedicated meeting as information becomes available. The updates include sharing of information and lessons learned from SRS, other DOE sites, and other published reports as part of the DOE technology development program. In these forums, the State of South Carolina has the opportunity to reveal expectations regarding technology and technology selection. Such revelations can be used to influence a particular technology path.

[10] S. R. Bush, *Tank Cleaning Technical Exchange 2006 Summary Report*, (CBU-PIT-2006-00067), p. 5.

[11] M. B. Birk (2011b), *loc. cit*.

4.0 WASTE REMOVAL PROCESS OVERVIEW

A tank is removed from service in seven sequential phases. The first five phases encompass the scope of the waste removal program as shown in Figure 4.0-1.

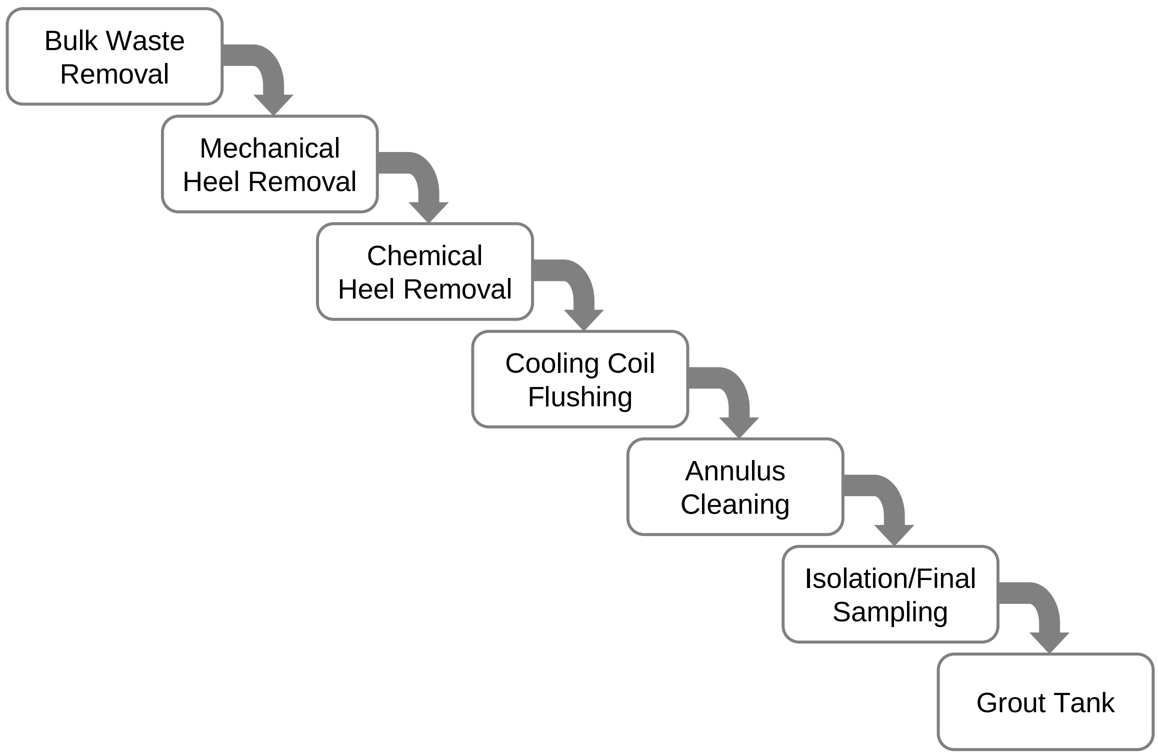

Figure 4.0-1. Waste Removal Program Phases

4.1 Bulk Waste Removal

The BWR phase extracts the majority of the tank waste, which reduces the radiological and environmental risk inherent in storing large volumes of waste material. The waste is removed at a rate to avoid disruption of feed to DWPF. The BWR phase places the waste tank in a condition where sufficient time can be given to the next two phases (the heel removal steps).

4.2 Mechanical Heel Removal

The heel removal phase is designed to leave the primary waste tank (i.e., the main storage vessel) ready to remove from service. Heel removal is normally accomplished in two steps. The mechanical portion involves non-chemical means such as mixing, spray washing and lancing. The mechanical methods help reduce the amount of chemicals used for the next step. Typically, the mechanical heel removal step continues until it is no longer effective.

4.3 Chemical Heel Removal

Chemical cleaning is the second part of heel removal and assists in removing waste constituents that could not be removed using mechanical methods. Integrating within the waste removal program, chemical cleaning techniques (i.e., acids, bases, oxidizers, surfactants, etc.) were first employed on Tank 16 in the early 1980s. The chemicals, and the products of reaction, must be compatible with downstream processes and equipment. Because of this, they are used judiciously and after much study.

4.4 Cooling Coil Flushing

Cooling coil flushing is designed to remove internal contamination that may have leaked into the coils after years of service. Some of the coils are broken; therefore, any water added to the waste tank from flushing would be removed before the grouting phase. Usually, flushing cooling coils is performed at the same time as heel removal because any flush water is routed to the primary waste tank.

4.5 Annulus Cleaning

Many of the older style waste tanks (Type I and II) have had waste solutions leak from the primary vessel into the annulus. Annulus cleaning for removal of contaminants involves rinsing the outside walls of the waste tank and the annulus pan. For the few waste tanks where more than a simple rinse is needed, mechanical means (e.g., mixing pumps or jets) may be used for the removal of residual waste. Annulus cleaning is done along with the primary tank heel removal activities (including chemical cleaning) because the rinse solution is sent to the primary waste tank, or another waste tank. The annulus is inspected before ceasing waste removal activities to confirm previously documented conditions, and to determine if additional leakage from the primary waste tank occurred during the waste removal process.

4.6 Isolation/Final Sampling

When a waste tank is cleaned and ready to remove from service, waste residue samples are taken from the primary tank and annulus, if applicable, to assist characterization of the remaining radiological and hazardous components. In addition, waste tank services and transfer lines are physically isolated preventing intentional or inadvertent waste tank operation.

4.7 Waste Tank Grout

Filling the waste tank with grout ensures final removal from service. Cement-based grout: 1) fills void spaces, 2) reduces the risk of subsidence, 3) provides a physical barrier from weather exposure, 4) is a physical deterrent to casual intruders, and 5) creates a condition that discourages transport of remaining residue.

5.0 WASTE REMOVAL TECHNICAL BASELINE OVERVIEW

This section describes the baseline equipment used, methods and techniques and the expected performance for the waste removal phases. Also included, is a discussion on the development path and selections on which the technologies are based. The baseline technologies are the end product of decades of research and operational experience. Systems engineering evaluations are used to evaluate various technologies that end up being components in the technical baseline. For example, a SEE conducted in 2003 formed the basis for a large portion of the waste removal program baseline.[12]

5.1 Bulk Waste Removal

5.1.1 Baseline Equipment

A jet mixing method is used during BWR to dislodge and suspend the settled sludge. The jets entrain and mix the surrounding fluid and the mechanical energy is supplied from pumps. Jet mixers are commonly used in large waste tanks where agitation with blade mixers is impractical. Jet mixing for BWR is provided by one of two devices, submersible mixer pump (SMP) or the standard slurry pump (SLP).

Submersible Mixer Pump: The SMP is a 7,600-gpm pump driven by a closed coupled 300 hp electric motor. Curtiss-Wright Electro-Mechanical Corporation specifically designed and manufactured the pump for utilization in SRS waste tanks. The SMP acquires suction from the bottom and pumps some of the slurry through cooling chambers in the motor before discharging out through two diametrically opposed side nozzles machined in the pump-motor casing. The fluid, with high velocity, exits the nozzles that mixes and agitates the waste tank contents. The pump-motor assembly mounts at the end of a long mast and inserts through a waste tank riser. The entire assembly oscillates on a slewing gear allowing the jets to sweep horizontally across the waste tank. In Figure 5.1-1, a jet nozzle can be seen as a hole in the casing side. The SMPs must operate with higher waste tank liquid levels than the SLPs. Deep submergence provides adequate net positive suction head and prevents the submerged jets from causing undue wave action on the liquid surface.

[12] Zupon, *loc. cit.*

Figure 5.1-1. SMP base during pre-installation testing

Standard Slurry Pump: The older generation SLP still in use employs a design developed by Savannah River National Laboratory (SRNL) in the 1970s (the waste tank end is shown in Figure 5.1-2). The concept is similar to the SMPs except the motor is located above the waste tank top. The pump connects to the motor with a long shaft through a water-filled column. The column is pressurized to prevent the migration of contamination to travel up the spinning shaft and outside of containment.

Figure 5.1-2. An SLP staged for trial runs at the SRS test facility

5.1.2 Baseline Methods and Techniques

In summary, sludge waste is removed by first agitating the waste tank contents with the mixer pumps, employing the principles from jet mixing theory. Three or four mixer pumps are installed through waste tank openings and spaced as equidistant as possible from each other to provide the largest coverage with minimal overlap. Sufficient liquid is added to the waste tank to suspend sludge solids. Usually, existing supernatant liquid from other waste tanks is used as the suspension medium to minimize introduction of new liquids into the tank farm system, however fresh water is occasionally applied. The water jets from the mixer pumps ablate the solids into a transportable suspension. The resultant suspension possesses non-Newtonian fluid behavior following the Bingham plastic flow model. The waste tank empties by transferring the suspension to another tank. A subsequent video inspection of the waste tank interior reveals the progress made by the mixer pumps, and if more solids need to be removed, the process is repeated until the removal goal is met. Equipment controls and monitoring are located nearby in a sheltered control center.

5.1.3 Baseline Performance

A mixer pump generates jet flow by acquiring suction from the pump bottom and discharging horizontally out of two diametrically opposed nozzles. The distance from the nozzle discharge to where sludge can be mobilized is called the effective cleaning radius (ECR). The ECR depends largely on the sludge tenacity and the time spent addressing the sludge layer. The pump bodies are rotated on a slewing gear at slow speeds (approximately 0.2 rpm) in a manner that causes the jet streams to sweep transversely over the vessel floor, thereby mobilizing waste across the effective radius of the jet streams. The ECR for the SMP is 52 feet, while the smaller SLP has an ECR of 32 feet. Within these radii, approximately 95% or more of the sludge is mobilized and removed. These radii can decrease depending on the type of sludge, interferences inside the waste tank, and the mixing duration.

5.1.4 Baseline Development Path

Limited waste tank access and numerous internal interferences led early SRS engineers to pursue jetting methods to agitate sludge (instead of the traditional mixing methods such as impellers and mixing blades). Sluicing was briefly investigated in the late 1950s. However, there was no driver to remove sludge from waste tanks until 1965, when increased canyon production, as part of the arms buildup of the 1960s, put a burden on waste tank space. During this time, the tank farms relied more on evaporation to create the compact waste form of saltcake. Therefore, sludge was removed and consolidated in a few waste tanks (e.g., Tanks 7 and 13) to make room for the saltcake. This effort started the technology development for BWR.

Cleaning Type I Tanks with High-Pressure Jets: Between 1967 and 1969 SRS removed the sludge from several Type I tanks using high-pressure water jets (3,000 psig) as seen in Figure 5.1-3. In the early 1970s, SRS began investigating the use of slurry pumps:

> *The removal of sludge is made difficult by a network of horizontal cooling coils located close to the bottom of the tanks… One promising method…is the use of high velocity jets*

of water to suspend the sludge for a sufficient time to allow the slurry to be pumped from the tank.[13]

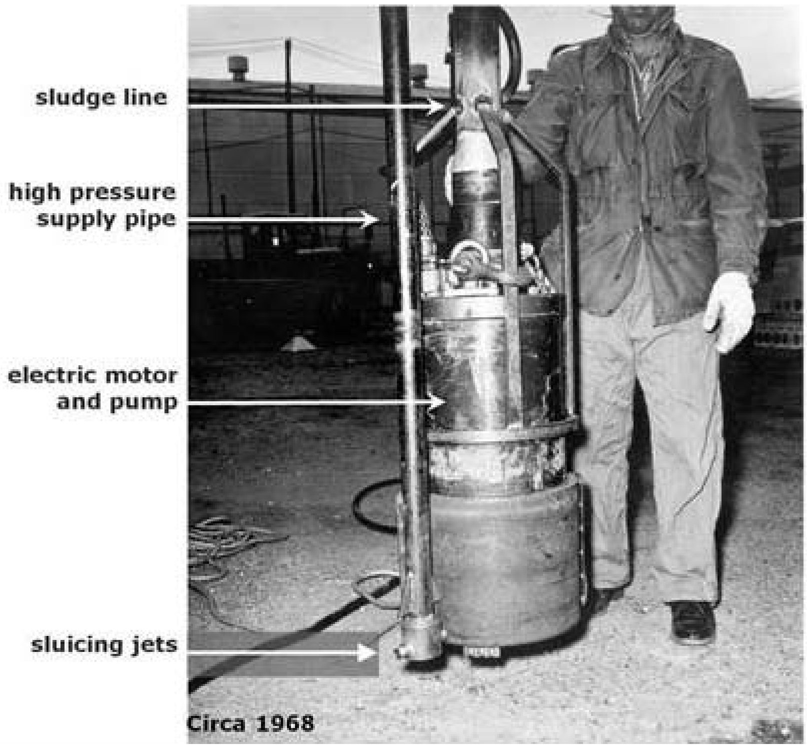

Figure 5.1-3. Type I waste tank sludge removal rig before being installed in the tank

The high velocity water jets dispersed the sludge into slurry that was removed using centrifugal transfer pumps. The opposing sluicing jets, mounted on a pipe mast, were rotated allowing the high velocity nozzles to sweep horizontally across the waste tank bottom. The transfer pumps, located next to the nozzle set, pumped out the slurry.

Rotation was provided by a rudimentary slewing gear that was located topside and driven by an electric motor connected with a bicycle chain. Each waste tank used six nozzle-pump assemblies. This early effort served as a basis for future BWR.

Although effective, the method used a lot of fresh water (approximately 5 gallons of water for every gallon of sludge removed). The first sludge removal program conducted over a 2.5-year period filled the tank farms to capacity with jet water and took a few years for the extra volume to be reduced through evaporation. Figure 5.1-4 shows the high pressure supply line rotated and driven by an electric-driven chain with the safety guard removed for the photograph.

[13] A. J. Hill, *Removal of Sludge from High Activity Waste Tanks*, (DP-1093), p. 5.

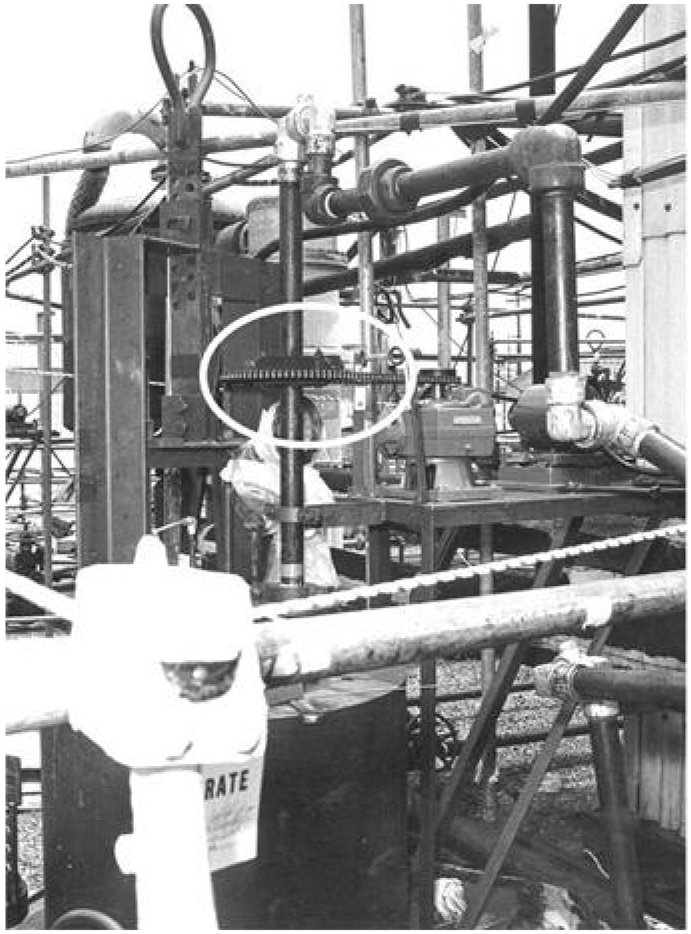

Figure 5.1-4: High-pressure line rotated with chain drive (circa 1968)

Standard Slurry Pump Development and Tank 16 Cleaning: In preparation for eventual closure of the tank farms, SRS began investigating ways to remove sludge without encumbering the available space. In 1977, SRNL completed development of a technique using a recirculation of supernatant liquid. The liquid resuspended and mixed the sludge eliminating the need to accommodate the extra volume. This method consisted of a single-stage centrifugal pump operating in the sludge with an operating pressure of approximately 100 psig. Recirculation liquid is drawn into the bottom of the pump and forced out through two oppositely directed nozzles. The resultant jetting action produced a sludge-slurry capability equal to that obtained with the high-pressure system. In addition to eliminating the large quantities of water, the low-pressure liquid recirculation technique required only 16.7% the power needed by the high-pressure system. The first successful test runs of a slurry pump were conducted in December 1978 and January 1979 in Tank 16. The test involved running a single Bingham-Willamette pump (now Sulzer Pumps) installed in Riser 2 for a total of 294 hours of mixing. Over 19,000 gallons of the original 77,000 gallons of settled sludge was mobilized and transferred. Almost 22,000 gallons of seal water (approximately 1 gpm) and

29,500 gallons of supernatant liquid (from Tank 23) were added to the waste tank. The pumps obtained a 30-foot ECR. In January 1979 and February 1979, two more slurry pumps were started totaling three pumps in Tank 16. The purpose of the multi-pump test was to extrapolate the number of pumps required to mix a waste tank. From the test results, three or four pumps were needed (seven to nine pumps were needed based on bench scale studies). Based on the test results, the slurry pump was a satisfactory candidate for use in the waste removal program. The researchers advised improvements in the pump design to reduce seal leakage and to facilitate decontamination.

Standard Slurry Pumps Continued Use: Since 1979, 16 waste tanks have used standard slurry pumps or some variation thereof for sludge removal, salt dissolution, and sludge mixing for DWPF feed preparation. The SLPs were first provided by Sulzer Pumps, followed by Lawrence Pumps. The second manufacturer provided SRS with a competitive alternative supplier. However, despite gradual improvements in manufacturing, SLPs demonstrated short operating life spans of 1,000 hours or less (approximately 42 days of continuous operation). The reliability problems were seal and bearing failures, which were caused by vibrations of the long shafts in the pump. The high cost of each pump (approximately $1 million), coupled with the cost of expensive waste tank top modifications, and the operation of a separate support system (bearing water) persuaded SRS to investigate alternative technologies. By 1996, a task team was formed to look for alternate ways to mobilize and remove waste from a waste tank, which ultimately led to the SMP development years later. SRS, however, remained committed to the jet mixing methods provided by the SLP. By 2001, SRS engineers made several design improvements to extend operating life. Using such improvements as fluidic bearings and seals, operating life extended up to a year.

Submersible Mixer Pump: Encouraged by SLP design improvement possibilities in conjunction with DOE sponsored alternative waste removal technologies, SRS engineers began investigating alternative mixer pump designs. In 1998, a submersible pump and motor design was proposed that would eliminate many of the vibration issues and the bearing water system. In 1999, funding through the DOE Office of Science and Technology enabled SMP development. The SMP is a "from the ground up" redesign of the mixer pump concept that can be inserted through a 2-foot riser opening and provide a large mixing capacity.

Submersible Mixer Pumps vs. Standard Slurry Pump: The SMP, together with the improved SLP, affords SRS with two viable jet-mixing options. They are used together as part of the waste removal strategy. The SMP is a more powerful mixer and is used frequently for rapid bulk sludge removal. The SMPs require more liquid in the waste tank for operation and they also add significant pump heat to the waste when operating. The SLP, although less powerful, can operate at lower waste levels, making them well suited for heel removal operations.

5.1.5 Baseline Technology Selection Methods

The SMP and SLP were chosen as the key equipment for BWR after several selection evaluations were performed. These include the following SEEs using the AHP:

- Tank 19 Waste Removal (1998)[14]

[14] N. R. Davis, *loc. cit.*

- Tank 18 Waste Removal (2001)[15]
- Waste Removal Balance of Program (2003)[16]

These evaluations reviewed the effectiveness, cost, and safety of the equipment while comparing them with other technologies available including in-house and external vendor designs. The SMP and SLP represent the state-of-the-art of this component type in this application.

5.2 Mechanical Heel Removal

5.2.1 Baseline Equipment

The mechanical heel removal step uses several methods to remove the stubborn accumulations of sediment remaining after a BWR campaign.

Waste Tanks with Cooling Coils: The same mixer pumps used for BWR continue to agitate a waste tank to enable additional heel removal. This is augmented using a *mechanical feed-and-bleed* (MFB) process, which extends the mixing time. The equipment for the MFB method is listed below:

- Downcomer (for fresh water addition)
- Temporary recirculation line (for continuous recirculated decant)
- Existing transfer lines and pumps (for batch recirculated decant)

The temporary recirculation line is shielded with lead blankets to reduce personnel exposure. The MFB method is described in more detail in Section 5.2.2.

Waste Tanks without Cooling Coils: The absence of internal interferences expands the type of cleaning tools that can be installed. In addition to the mixer pumps, robotic crawlers are available for vacuuming the waste tank bottom. The crawler is equipped with an eductor that is used to aspirate material off the floor and transfer the suspension to a nearby waste tank. The crawler is also equipped with forward sprays to dislodge and suspend hardened sediment. The rig is controlled by an operator using hydraulic controls located near the waste tank. The illustration in Figure 5.2-1 shows the Mantis platform.

[15] Abell, *loc. cit.*
[16] Zupon, *loc. cit.*

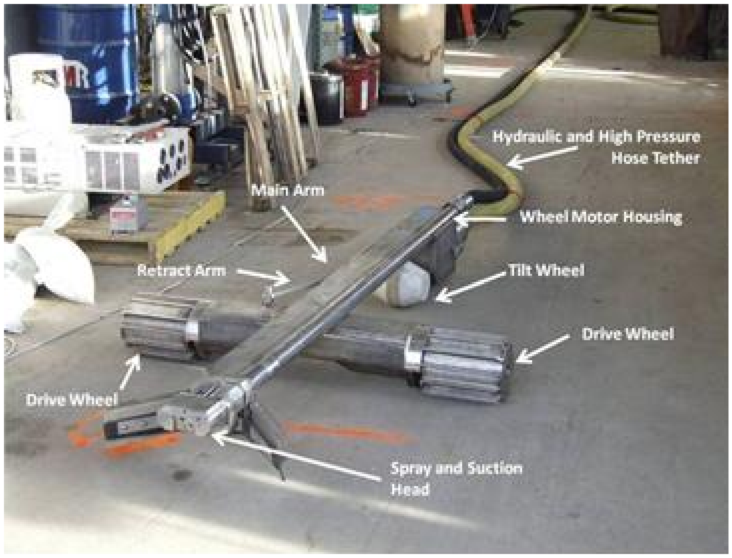

Figure 5.2-1. Mantis platform in the vendor's test facility

5.2.2 Baseline Methods and Techniques

Waste Tanks with Cooling Coils: MFB uses waste tank agitation and water makeup. Removal is accomplished by agitating the contents using three or four mixer pumps while simultaneously pumping the sludge suspension to a destination waste tank. Makeup water is supplied at the same rate as the transfer rate to maintain the source waste tank level thus extending the mixing time. The makeup water can either be fresh water supplied through an installed downcomer or recirculated liquid supplied through a dedicated return line from the destination waste tank. For the latter case, the destination waste tank serves as a settling basin allowing the particles to sink. A temporary waste transfer and pumping system decants the clarified portion of the supernatant liquid back to the source waste tank. Refer to Figure 5.2-2 for a schematic of this method. Both of these arrangements maximize mixing time, which allows more time for material to be removed. The dedicated transfer line method offers the benefit of not adding water volume to the tank farms. Similar results are achieved using the existing transfer system. This involves pumping the contents of the source waste tank down to a minimum level while maintaining mixer pump operation. After sufficient settling, the same liquid volume is returned from the destination waste tank back to the source waste tank. Figure 5.2-3 shows the temporary recirculation line installed above-grade that was used to clean a Type I tank.

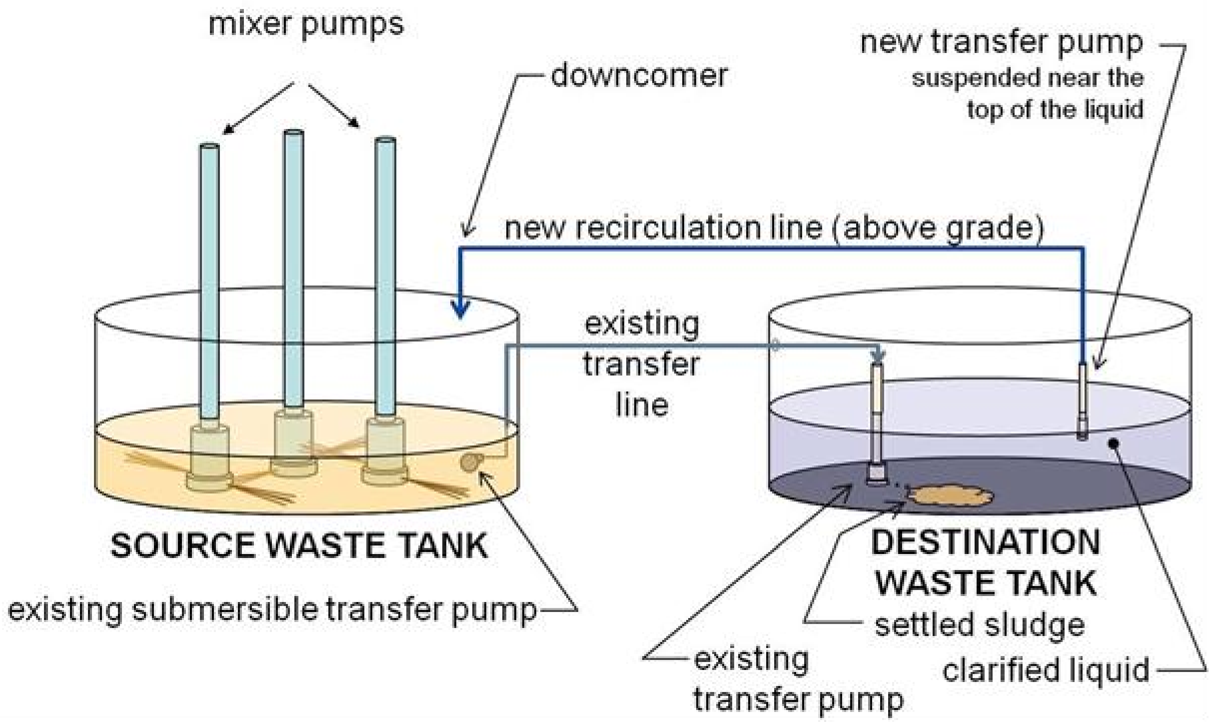

Figure 5.2-2. Recirculation Heel Removal Method

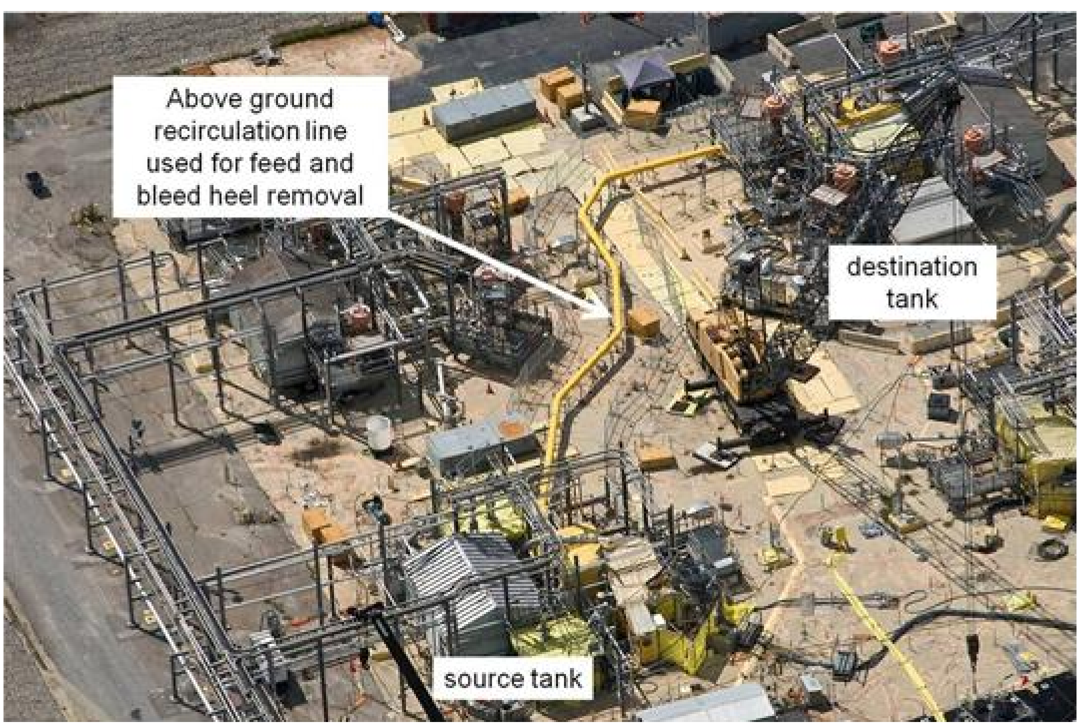

Figure 5.2-3. Above-grade shielded line transfers clarified liquid (2010)

These techniques are adaptations of heel removal methods found in the chemical process industry and in storage tanker cleaning. Commercial methods encourage the reuse of liquid and the minimization of secondary waste streams. Other tools, such as lances and sluicers, are sometimes employed to dislodge hard mounds or hard-to-reach deposits. For example, a hydro-lance broke up hardened zeolite mounds in Tank 19 (Figure 5.2-4). These are used while the waste tank level is low when the waste can be accessed.

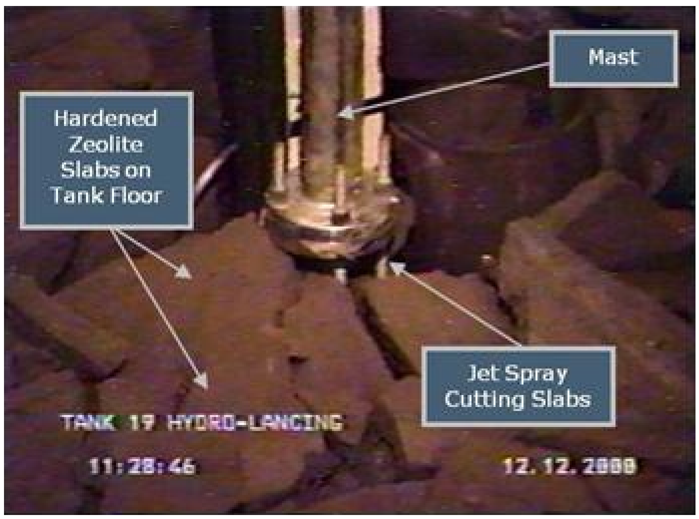

Figure 5.2-4. Tank 19 zeolite blocks being broken up by a high pressure hydro-lance

Another technique is to direct the nozzles of the mixer pumps towards a mound and leave the mixer in a stationary position for an extended time (indexing method). Experience has shown that mounds formerly outside mixer pump reach can be reduced using this technique. The time needed to make this method effective is substantially longer than that normally used for routine BWR.[17]

Waste Tanks without Cooling Coils: The mixer pumps heel removal method can also be employed for waste tanks without cooling coils, however, the open architecture allows for more options. The volume of the heel in Tanks 18 and 19 was reduced using a remote controlled vacuum cleaner (Mantis) that crawled across the bottom aspirating and transporting the waste material (Section 5.2). To break up solidified accumulations forward water sprays were attached to the crawler. The crawler motion was controlled by a nearby operator using a multi-function joystick while observing the movement on a video screen.

5.2.3 Baseline Performance

The purpose of mechanical heel removal is to remove the maximum waste possible, thereby reducing, or eliminating the chemicals necessary in the chemical cleaning step. Only a thin layer of residue remains on areas of the waste tank floor within the mixer pump ECR. The Mantis performance in Type IV tanks is dependent on the physical properties of the waste.

[17] B. A. Hamm, W. L. West, and G. B. Tatterson, "Sludge Suspension in Waste Storage Tanks," *AIChE Journal*, August 1989, p. 1394.

Previous results demonstrated that sticky, mud-like sludge like that of Tank 18 is more difficult to remove than the grainy sand-like sludge found in Tank 19.[18]

5.2.4 Baseline Development Path

Technology development to remove waste tank heels began with Tank 16 in the early 1980's. Early methods required large volumes of water to remove the majority of the solids before chemical cleaning methods could be employed. Similar approaches (using large volumes of fresh water) were taken to reduce the heel volumes in Tanks 7, 8, 11, and 12. Before the Mantis method (Tank 19), multiple additions and transfers were conducted by necessity when reducing the volume of the zeolite mounds.

Mechanical Feed and Bleed Method: The MFB method is a relatively new development, but uses a proven concept. The method was first proposed in 2005 with a tabletop demonstration and was adopted as a viable heel removal method in September 2009.[19] The fresh water feed and bleed method was employed on Tank 5 in January 2010[20] and the continuous recirculation method was used for Tank 6 in July and August of 2010.[21] Batch recirculation was performed on Tank 4 between October 2010 and February 2011.[22]

Mantis Development: SRS hosted a waste tank cleaning technical exchange in Atlanta, Georgia, on March 27, 2006 and March 28, 2006, to facilitate the exchange and generation of innovative ideas on waste tank cleaning in preparation operational closure. The conference promoted a vigorous and constructive exchange. Suppliers attended the conference to learn about the DOE needs and constraints and to promote their ideas regarding radioactive waste tank cleaning. Following the technology exchange in Atlanta, SRS received unsolicited proposals from several vendors. One such proposal was a particularly noteworthy device. SRS representatives observed demonstrations of the device in operation at the vendor's facility during May 2006. The device employed a simple yet effective crawler system. The platform of the device is the same as a mechanism previously used in a tank at the Hanford Site. The vendor modified the platform incorporating a suction device designed to remove fast-settling particles (zeolite) and dense and cohesive material (sludge). The suction device itself is an eductor driven by ultra-high pressure water (greater than 10,000 psi). The eductor effectively demonstrated the aspiration of solids and transport distances sufficient to reach other receipt vessels. In addition, the vendor resolved a tether management issue that plagued other crawler systems. The SRS teamed with the vendor deploying the device in Tanks 18 and 19.[23] The time from project start to deployment was roughly 3 years.

5.2.5 Baseline Technology Selection Methods

The MFB method was selected with the SEE process. The Mantis was chosen from information gathered at a technical exchange.

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[18] T. B. Caldwell (2006), "Trip Report for Sand Mantis at the Hanford Cold Test Facility," Interoffice Memorandum to N. R. Davis, (LWO-CES-2006-00006), p. 2.
[19] Clendenen (2009), *op. cit.*, p. 10.
[20] G. B. Clendenen (2010), *Tank 5 Waste Removal Operating Plan*, (SRR-CES-2009-00022), pp. 63-71.
[21] T. B. Caldwell (2010a), G. B. Clendenen, and J. J. Purohit, *Tank 6F Phase II Mechanical Sludge Removal (MSR): A Heel Removal Technique using Agitation and Continuous Recirculation*, (SRR-CES-2010-00031).
[22] T. B. Caldwell (2010b), "Tank 4 Mixing Strategy to Support Sludge Batch 7B," Interoffice Memorandum to T. L. Williams, (SRR-LWE-2010-00350).
[23] Frank Fisher, *Sand Mantis System Proof of Concept, Phase 1 Demonstration Completion Report*, TMR Associates, LLC, Lakewood, CO, 2007.

MFB Method: A modified SEE was completed in the fall of 2009 to address the heel left behind in Tanks 5 and 6 after a chemical cleaning campaign. Using AHP, the evaluation team proposed variations of the MFB method.

Mantis Method: The Mantis was selected from a field of concepts that were presented at the Atlanta technology exchange after consultation with a team of subject matter experts. The Mantis, at that time, was deemed to be the most practical and technically mature of the reviewed options.

5.3 Chemical Heel Removal

5.3.1 Baseline Equipment

Tanker trucks added chemicals to the waste tanks through downcomers or spray wash nozzles. The spray wash nozzles permit the chemicals to cover the walls and cooling coils. Mixer pumps (SMP or SLP) provide the mixing. The standard chemical used is oxalic acid (OA) in strengths from 1 to 8 wt%. The Enhanced Chemical Cleaning (ECC) process, which is currently under development, is investigating lower acid strengths (e.g., approximately 2 wt%). Other chemicals (such as sulfuric and nitric acid) are also being researched for possible improvement to OA performance.[24]

5.3.2 Baseline Methods and Techniques

Chemical cleaning employs OA to reduce particle size, dissolve or otherwise loosen solids not removed by mechanical methods and water addition alone. The OA is pumped or sprayed into the waste tank to clean contaminants from the internal waste tank surfaces (e.g., walls, cooling coils, support columns, equipment). Approximately 150,000 gallons of OA solution added to the waste tank submerges the mixer pumps and facilitates agitation. The mixer pumps provide agitation and heat to accelerate the chemical reaction and to suspend loosened solids. After several days of continuous agitation, the spent solution is pumped to a waste receipt tank. The receipt waste tank holds excess sodium hydroxide (from a previous addition) to neutralize the low-pH spent solution, thus maintaining corrosion control. Several (typically three) of these evolutions ("strikes") are performed. At the conclusion of chemical cleaning campaign, the waste tank interior is washed with water to rinse the acid from internal surfaces and further dislodge loose contamination. This rinse is pumped out leaving the waste tank empty for subsequent remediation.

5.3.3 Baseline Performance

One of the strongest organic acids available for use in waste tank cleaning is OA. As discussed later, mineral acids such as hydrochloric or sulfuric have a tendency to be overly aggressive, producing excessive amounts of flammable gasses when reacting. Stronger organic acids have a propensity to be toxic, explosive, or possess violent reactivity. The OA does not dissolve all of the sludge even with the dissolution rate improvement that mixer pump agitation provides. It may take several strikes to maximize the benefit of OA.[25]

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[24] W. D. King and M. S. Hay, *loc. cit.*
[25] R. N. Vemulapalli, *Tanks 5 and 6 Oxalic Acid Aided Heel Removal Flowsheet*, (LWO-PIT-2006-00066, Rev. 2), p. 7, Sect. 3 "Approach."

5.3.4 Baseline Development Path

The development of heel removal techniques using chemicals is straightforward. In the 1970's SRS chose OA over other chemicals, including Ethylenediaminetetraacetic Acid (EDTA) and sulfamic acid, for several reasons:[26]

- **Strength -** OA is over 3,000 times stronger than acetic acid.[27] Mineral acids such as hydrochloric or sulfuric have the tendency to attack the waste tank metal and produce excessive amounts of flammable gasses when reacting. Stronger organic acids are inclined to be toxic (trichloroacetic acid),[28] explosive (picric acid),[29] or possess violent reactivity (benzenesulfonic acid).[30]
- **Safe -** OA is an attractive cleaning agent because the corrosion damage to the carbon steel waste tanks is limited through the passivation of the metal with the formation of iron (II) oxalate.[31] Oxalic acid is safely handled using appropriate personal protection equipment.[32]
- **Available -** The reagent is commercially available from several sources.
- **Compatible with long-term lay-up -** The oxalates formed from the acid-base reaction are also reducing agents, which favor closure chemistry.
- **Effective -** OA dissolves most iron compounds, some aluminum compounds, and some uranium compounds. The acid works as a dispersant by reducing the size of other solids. Sludge that has reacted with OA is easier to mobilize and transport out of a waste tank.[33]

On the down side, laboratory studies show that nickel-based sludge is unaffected by OA.[34] Moreover, plutonium-based solids are resistant to OA attack.[35] This would imply that using OA would preferentially dissolve the clean compounds, leaving behind some contaminants (e.g., plutonium). This was a concern with SRS engineers when OA was first proposed for Tank 16 heel removal in 1978.[36] However, sampling performed after completion of chemical cleaning on Tank 16 did not indicate a separation of components and researchers confirmed the mixer pumps effectively suspended the sludge that was treated with OA.[37]

Two challenges working with OA are described below:

- **Byproducts -** The products are toxic to humans. The conjugate base is an oxalate ($C_2O_4^{-2}$) with sodium oxalate ($Na_2C_2O_4$) being the most common salt formed during

[26] R. F. Bradley and A. J. Hill, *Chemical Dissolving of Sludge from a High Level Waste Tank at the Savannah River Plant*, (DP-1471), p. 18.
[27] John A. Dean, *Lange's Handbook of Chemistry*, Table 8.8, "pK_a Values of Organic Materials in Water at 25°C", pp. 8.24 and 8.63. Solving for the acidic dissociation constant (K_a) using pK_a = -log$_{10}$ K_a by obtaining the pK_a values for acetic acid and oxalic acid from the table cited.
[28] *Trichloroacetic acid*, CAS No. 76-03-9, Material Safety Data Sheet, p. 1.
[29] *Picric acid*, CAS No. 88-89-1. Material Safety Data Sheet, p. 2.
[30] *Benzesulfamic acid*, CAS No. 98-11-3, Material Safety Data Sheet, p. 1.
[31] M. E. Stallings, D. T. Hobbs, and B. J. Wiersma, Dissolution of Simulated and Radioactive Savannah River Site High-Level Waste Sludges with Oxalic Acid and Citric Acid Solutions, (WSRC-TR-2004-00043), p. 40.
[32] *Oxalic acid*, CAS No. 144-62-7, Material Safety Data Sheet, p. 3.
[33] W. L. West, "Tank 16 Demonstration Water Wash and Chemical Cleaning Results," Interoffice Memorandum to O. M. Morris, (DP-80-17-23), p. 1.
[34] M. E. Stallings, D. T. Hobbs, and B. J. Wiersma, *op. cit.*, p. 27.
[35] D. G. Karraker, *Solubility of Uranium, Plutonium, Iron and Manganese in Weak Nitric Acid and Oxalic Acid Solutions: A Literature Survey*, (WSRC-RP-98-00091), p. 1.
[36] J. L. Forstner, "Nuclear Safety Considerations during Sludge Removal from Tank 16 (Supplement 1)," Letter to E. S. Bridges (DPSPU-78-272-33), p. 1.
[37] J. R. Fowler, "Radiochemical Analyses of Samples from Tank 16 Cleanout," Interoffice Memorandum to R. B. Ferguson (DPST-81-441), p. 1.

waste tank cleaning. However, the correct personal protection equipment will safeguard workers when handling this material. Furthermore, the protection measures taken when working in a radiological environmental sufficiently protects the workers from the chemical hazards.

The oxalates also have limited solubility in solutions with high sodium and hydroxide concentrations. This means that some of the products are temporarily soluble. To counteract this effect, the solutions are removed from the waste tank before the compounds have a chance to precipitate.

- **Dissolution Effects -** The second programmatic issue is the preferential dissolution of some compounds over the others. Presently, no single acid has been identified that will dissolve all of the sludge compounds equally while not also dissolving the waste tank. However, OA is the most "complete" chemical for dissolving most of the compounds (while protecting the waste tank), and aiding in the dispersion and suspension of the remaining compounds.

Technology development continues to improve chemical cleaning methods. The ECC is an OA process, under development, that destroys or oxidizes the oxalates before introduction to the destination waste tank. The process is accomplished through a separate oxidation process. The prototype waste tank for ECC will be Tank 8. The SRNL is researching possible augmentation or replacement of OA to improve overall removal rates. The SRNL investigated improving dissolution performance using supplemental acids mixed with traditional OA solutions. However, further testing, analyses, and safety evaluations are needed before being considered as part of the chemical heel removal baseline. The SRNL found that an OA-nitric acid mixture or an OA-sulfuric acid mixture dissolves iron-based sludge more completely than OA alone. They also discovered the aluminum-based components in the sludge dissolves better with strong solutions of sodium hydroxide.[38]

Appendix A provides a timeline of the chemical cleaning heel removal development at SRS.

5.3.5 Baseline Technology Selection Methods

In 2003, a Heel Removal Task Team was formed to investigate the best method to dissolve waste tank heels, and to implement the method into the modern LW program. The team concluded that OA to be the safest and most effective cleaning agent.[39]

Three more alternative evaluations concluded OA as the preferred component in chemical heel removal. The Waste Removal Balance of Program (2003) SEE[40] selected OA as the preferred candidate for chemical cleaning. In 2007, a method of inventive problem solving identified an enhanced process using OA, which formed the basis for ECC.[41] In 2009, a SEE investigated multiple chemical cleaning regimens, which included OA. The OA plus nitric acid scored highest with OA (baseline reagent) coming in second. Additional testing,

[38] W. D. King and M. S. Hay, *Alternative and Enhanced Chemical Cleaning: Basic Studies Results FY2010*, (SRNL-STI-2010-00541), p. 60.
[39] M. J. Barnes, *Waste Tank Heel Chemical Cleaning Summary*, (WSRC-TR-2003-00401), p. 70.
[40] Zupon, *loc. cit.*
[41] E. T. Ketusky, *Determination of an Alternative Technology for HLW Tank Chemical Cleaning*, (WSRC-STI-2007-00587), p. 11.

analyses, and safety evaluations are needed before OA supplemented with another reagent can be used in the waste tanks.[42]

5.4 Cooling Coil Flushing

5.4.1 Baseline Equipment

Cooling coil flushing involves flushing the residue from the interior surfaces of the cooling coils with clean water using standard pipe flushing methods. The equipment used is commercial-grade water connections and hoses.

5.4.2 Baseline Methods and Techniques

Flushing the cooling coil assembly involves connecting a clean water supply to each coil circuit by first isolating the circuit and removing pipe spool pieces located in the valve house (above grade) so that connections can be made. Fresh water is provided through hoses that are connected to the circuits using quick disconnects. Water is allowed to flow through the pipes (cooling coil) for at least a three-volume flush. The slightly contaminated rinse water is collected in a tanker vessel or routed to another waste tank. When there are broken coils, the flush water drains in to the waste tank where it is later collected and pumped to another waste tank.

5.4.3 Baseline Performance

A three-volume flush is expected to remove greater than 99% of the loose contamination in the coiling coils. This is the flush volume traditionally used for flushing the SRS waste transfer lines.[43]

5.4.4 Baseline Development Path

Flushing cooling coils and rinsing the annulus are straightforward solutions that did not warrant a protracted development process. This process is considered a routine decontamination method employed by nuclear industry.[44]

5.4.5 Baseline Technology Selection Methods

The Waste Removal Balance of Program (2003) SEE[45] selected these methods as the preferred candidates for cooling coil flushing and annulus cleaning.

5.5 Annulus Cleaning

5.5.1 Baseline Equipment

Clean water sprayed on the primary liner (waste tank wall) is the baseline for removing small salt deposits on the wall and annulus floor. A positive displacement pump, installed through one of the 2-foot diameter access openings (on the waste tank top), removed the liquid from the annulus to the primary waste tank leaving the annulus in a near-dry condition.[46] A

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[42] C. J. Martino, "Evaluation of Alternative Chemical Cleaning and Enhanced Chemical Cleaning Methods," Interoffice Memorandum to S. L. Marra, (SRNL-L3100-2009-00118), p. 14.

[43] N. R. Pasala, *Tank Farm Transfer Control Program & Pump Tank Transfer Jet Control Program*, (WSRC-TR-2002-00403), p. 35.

[44] Electric Power Research Institute, *A Review of Plant Decontamination Methods 1988 Update*, (EPRI NP-6169), p. 2-13.

[45] Zupon, *loc. cit.*

[46] Rudy Jolly, "Path Forward – Tank 5 & Tank 6 Annulus Cleaning Methodology", Interoffice Memorandum to Mssrs. R. Boisvert, John Bennett, and David Little, (LWO-LWE-2008-00018), p. 3.

commercially available crawler (Force Institute Commercial Wall Crawler[47]) assisted in removing deposits in Tanks 5 and 6. The small number and size of the salt deposits permitted effective use of the crawler.[48] Large deposits, such as those found in Tank 16, will require another method of removal, such as robotic manipulator arm.[49]

5.5.2 Baseline Methods and Techniques

The small wall cracks observed in some Type I and II tanks only allow soluble liquid components to leak from the primary liner to the secondary liner. The cracks are too small for solid particles to pass through. Therefore, the soluble waste in the annulus can be washed off with clean water. Annulus cleaning involves spraying water to the old leak sites using the wall crawler. Water is allowed to drain to the annulus bottom. A simple spray wand is extended down in the annulus for easy to reach spots. After the spot cleaning, fresh water is poured into the annulus pan. After the soak period, the rinse is pumped to the primary waste tank or to another waste tank. Waste tanks with large volumes of soluble material require the process to be repeated until the salt is dissolved and transferred out.

5.5.3 Baseline Performance

The expectation is that most of the soluble species will be removed from the annulus.[50] Using Tanks 5 and 6 as a basis, the goal is to not require further washes after the first water rinse.[51]

5.5.4 Baseline Development Path

A previously successful wall crawling platform was adapted for cleaning use. The crawler is a modification of a Force Institute commercial wall crawler used to perform camera inspections in the annulus.

5.5.5 Baseline Technology Selection Methods

The methods used for annulus cleaning are a continuation of past practices. The functional design criteria for DOE Project S-2081 and the functional performance requirements for Project S-W183 describe the technical requirements and methods for annulus cleaning.[52, 53]

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[47] R. L. Minichan, R. E. Eibling, J. B. Elder, K. E. Kane, D. Krementz, R. W. Vandekamp, and N. J. Vrettos, *Annulus Closure Technology Development Inspection / Salt Deposit Cleaning Magnetic Wall Crawler*, (WSRC-STI-2008-00308), p. 4.
[48] Rudy Jolly, *op. cit.*, p. 2.
[49] George R. Davis, *Scope of Work and Strategy, Tank 16 Annulus Cleaning*, (SRR-LWE-2010-00261), pp. 4-5.
[50] B. A. Martin, "Tank Closure Inputs and Assumptions in Support of the Liquid Waste Disposition Processing Plan, Revision 1," Interoffice Memorandum to P. J. Hill (CBU-PIT-2006-0047, Rev. 2), p. 3.
[51] Nilesh P. Badheka and Nader S. Elraheb, *Annulus Cleaning Technical Plan* (CBU-LTS-2004-00109), p. 7.
[52] S. C. Lee, *Functional Design Criteria Waste Removal and Extended Sludge Processing*, (G-FDC-G-00029).
[53] T. A. White, *Functional Performance Requirements Project S-183*, (FPR-G-00019).

6.0 BASELINE ALTERNATIVES

The original slurry pumps were successful in removing the bulk of the sludge and reducing the heel to acceptable levels. However, they had an operating life of roughly 1,000 hours (42 days of continuous operations). Experience gained from cleaning Tank 16 demonstrated that a longer operating life (in excess of 1,000 hours) for remediation tools is influential for them to be economical. The installed cost of a single pump exceeded $1 million (not including disposal fees). In the mid 1990s, the high cost and short service life of these pumps led SRS to explore baseline alternatives. The evaluation effort eventually expanded to include looking at alternative technologies other than mixing, which included sluicing, dredging, robotics, and other non-standard methods. This section summarizes the efforts of these studies.

6.1 Tanks Focus Area

The Tanks Focus Area (TFA) was initiated in 1994 to serve as the DOE Office of Environmental Management's national technology development program for radioactive waste tank remediation. The national program formed to increase integration and realize greater benefits from DOE's technology development budget. The TFA managed, coordinated, and leveraged technology development in support of the requirements of DOE's five major waste tank sites (SRS, Hanford Site, Idaho National Engineering and Environmental Laboratory, Oak Ridge Reservation, and West Valley Demonstration Project). The program lasted through 2004.

By 1996, DOE sponsored new ways to remediate a waste tank, in part through the TFA and through other contractual initiatives. The drivers were several-fold:

- Reduce reliance on the SLPs
- Reduce remediation costs and improve on closure schedules by using collective knowledge and methods common to waste tank remediation
- Improve remediation effectiveness by leveraging technologies (and lessons learned) from within the DOE complex and commercial remediation enterprises

6.2 Alternative Waste Removal Task Team

In 1996, SRS initiated an effort to investigate waste removal alternatives in conjunction with the TFA process. The Alternative Waste Removal Task Team was formed with the specific goal to "deliver alternative waste removal techniques to management for the purpose of retaining funding for field implementation." The team, consisting of several subject matter experts, brainstormed, researched, and investigated various sludge removal and agitation techniques (an alternate to SLPs). The following mechanical (e.g., non-chemical) means were investigated:

- Slow erosion technique (dendritic flow)
- Dredging (air lifts)
- Sluicing
- Free jet flow agitators (electric motor-driven propellers)
- Pulse jet agitators
- Air pulse agitation
- Steam jet agitation
- Multiple mini mixers
- External agitator blades

- Submersible mixer pumps
- Robotic manipulator arms
- Crawlers
- In-tank vacuum aspirators

Team members investigated each of these options, sponsored in part by the TFA.

6.3 Alternative Tank Cleaning Tools

The following summary describes the methods proposed by the Alternative Waste Removal Task Team that led to prototype development.

6.3.1 Free Jet Flow Agitators

During Tank 17 closure in 1997, a small, submerged agitator was installed in the waste tank before the liquid contents were removed. The purpose was to agitate sludge fines promoting transfer to an adjacent waste tank. The agitators (Flygt mixers), manufactured by ITT Corporation, are commonly used to suspend municipal sludge in wastewater treatment plants (Figure 6.3-1). The effect of the mixer was not apparent, but the tank liquid clouded when the mixer was started providing sufficient justification to pursue this mixer type for further study. In 1998, mixing studies began on Tank 19 (a tank holding roughly 33,000 gallons of fast-settling zeolite particles).

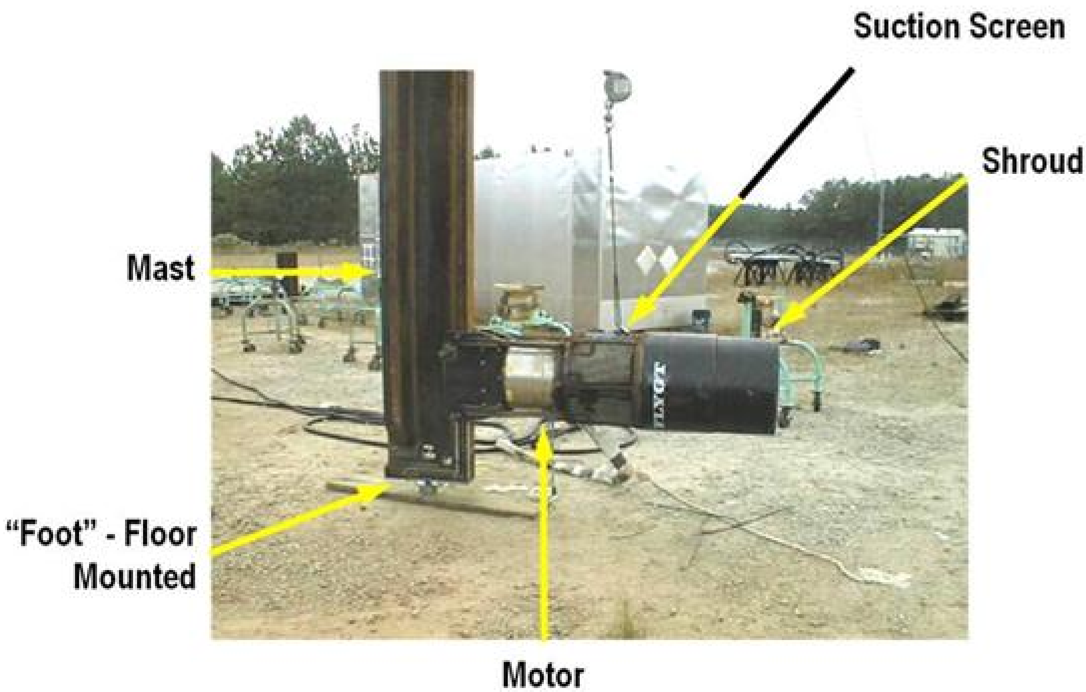

Figure 6.3-1. Tank 19 Flygt mixer at the SRS test facility

The Pacific Northwest National Laboratory, SRNL, and SRS sponsored waste tank mixing studies using three Flygt mixers mounted on a vertical mast with the flow direction aimed horizontally. The studies recommended rotation of a mounting mast for increased mixing success. After considerable development, three of these mixers were installed in Tank 19 for sludge and zeolite mixing. Consistent with first-of-a-kind developmental efforts, several technical issues arose that reduced the efficacy of the method. First, the diameter of the mixer blades had to be cut down to fit through the 24-inch tank opening reducing the hydraulic power of each mixer. Second, the mixers did not exhibit the long-term durability realized in commercial applications (one of the mixers failed shortly after start up). Third, the zeolite layer proved more tenacious than the surrogate material used during mock up testing. However, several lessons were learned from the experience that carried through to future technology development efforts:

- Mock up testing (including surrogates) must emulate field conditions.
- Commercial technologies require significant development to be "tank ready” for waste removal. The cost savings of using an off-the-shelf component is often overtaken by the expense to adapt the component for a HLW tank. The mast assembly cost for each Flygt mixer was comparable to the SLP.
- Development or adaptation of one technology sometimes provides use in later applications. The oscillating slewing gear to turn the mixers were later adapted for the SMP. In addition, the mixer's floor-mounted foot support was found of use with the SMPs.

6.3.2 Sluicing

A commercial fire monitor by Akron Brass Company was installed in Tank 17 to provide a jet sluicing spray (directed spray nozzle) to facilitate residual heel removal. The monitor used a small control box, located remotely above the waste tank top, to pan and tilt. The monitor, known as the water brush (named for its ability to "sweep” the waste tank floor), swept sludge accumulations towards the suction of the temporary transfer pump. Two steel disks called spray shields, prevented water spray from exiting the waste tank (Figure 6.3-2). The observation concluded that the sluicing process had a tendency to wash away fines, leaving behind larger particle material. The Hanford Site sluicing operations encountered the same phenomenon.

Figure 6.3-2. Water brush during fabrication

6.3.3 Robotic Manipulator Arms

A prototype arm was developed for Tank 19 by Delphinus, a technology company, specializing in robotic manipulators, but the arm was never deployed because of ineffectiveness (as evidenced by trial runs) and cost. Other manufacturers were pursued, including PaR Systems and GreyPilgrim, but not selected. The marginal benefits of the one-of-a-kind devices represented by these vendors did not justify the development and deployment cost. Cooling coils exist in most of the waste tanks and robotic manipulator arms at that time did not have the sophistication to maneuver around the obstructions in a Type I, II, or III/IIIA waste tank.

6.3.4 Advanced Design Mixer Pump

The advanced design mixer pump (ADMP) was originally conceived by the Hanford Site and supported by SRS to provide a more reliable and maintainable mixer pump for use throughout the DOE complex. Supported by the TFA, the ADMP underwent an extensive test program at SRS between 1998 and 2002 to assess reliability and hydraulic performance. The ADMP was a departure from the SLP design. Like the SLP, the ADMP was a long-shaft, vertical, centrifugal mixer pump with two tangential nozzles. However, the column was filled with gas, instead of liquid, the bearings were oil lubricated, and the ADMP is bigger than the SLP. The pump is 55 feet long, with a 16-inch column, 39-inch casing, 18-inch mixed-flow impeller, two 6-inch diameter nozzles, and a 300 hp motor. The ADMP did not fit through a 2-foot opening, but did fit through a FTF Type IV tank center riser. The pump flow rate was 10,400 gpm at 1,185 rpm with 52 feet of head. The theoretical cleaning radius was over 50 feet. However, despite the horsepower, the mixer pump underperformed when used in Tank 18. The large size precluded continued interest for Type I tank remediation.

6.3.5 Pulse Jet Mixer Agitation

The pulse jet mixer is a device developed and marketed by NuVision and employs a pair of compressed air powered pulse jets. During the charge phase, vacuum is pulled on the charge vessel thereby filling the vessel through the jet nozzle with liquid from the waste tank. During the compression phase, the liquid is forced out of the vessel under air pressure at a high rate of volume. The elapsed time of one charge and compression cycle varies, but is roughly 1 to 2 minutes. As with the SLP, the assembly is rotated or oscillated using a turntable mounted above the waste tank opening. A system was eventually designed and developed for Tank 24, but not installed because of air contamination concerns.

7.0 SUMMARY

The SRS has applied the knowledge learned over the past 40 years to optimize the technology development process. Even with due confidence that the waste removal technical baseline is well defined and successful, existing tools and agreements continually assist improvement or optimization of the cleaning method learning process. Some examples of current efforts include:

- Developing new mixer pumps for optimized agitation and multipurpose use based on lessons learned from SLPs and SMPs
- Working with other companies developing methods to remove sludge located directly under risers of waste tanks with cooling coils
- Monitoring technology progress at other DOE sites (e.g., Mobile Arm Retrieval System, the advanced reach sluicing system at the Hanford Site)
- Monitoring the mixed acid flow sheet development (e.g., OA, nitric acid) funded by the DOE technology development program
- Continuing ECC process development
- Continuing the small crawler vacuum cleaning system development, based on lessons learned from the Tanks 18 and 19 crawler

The actual process used for waste removal varies depending on the waste tank service history, the remaining waste physical characteristics, the waste tank system physical configuration, and the waste removal timing. In summary, SRS has a waste tank cleaning process that works with greater than 99% of waste removed.[54, 55, 56] Also in place is a robust, rigorous technology selection process to evaluate new technologies as they mature. To date, there is an extensive list of technology options that have been evaluated. The technologies that were selected have been optimized to address physical, chemical, and system challenges. Future improvements are likely to be incremental since current processes are removing greater than 99% of original waste volume.

[54] B. A. Stephens, *Tank 6 History of Waste Removal 1964 through 2010*, (SRR-CWDA-2011-00005, Rev. 1), 2011.
[55] B. A. Stephens and K. D. Gilbreath, *Tank 5 History of Waste Removal 1959 through 2010*, (SRR-CWDA-2011-00033, Rev. 1), 2011.
[56] M. B. Birk (2011a), *loc. cit.*

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APPENDIX A: Chemical Heel Removal Development Timeline

APPENDIX A

The progressive development of chemical cleaning for heel removal timeline at SRS is shown below.

Year	Development Description
1977	OA was first proposed by SRNL. Other chemicals reviewed included EDTA, sulfamic acid, glycolic acid, citric acid, and sulfuric acid. Acid combinations were studied for effectiveness.
1981	Tank 16 was cleaned using hot OA at 4 wt% leaving behind only surface residue of iron oxalate.
1981	OA and hydrogen peroxide were studied for future waste tank cleaning improvement. The combination was more effective than OA alone, except the peroxide-OA cleaning solution was highly corrosive to the waste tank, and the dissolution process produced measurable quantities of flammable gas.
1981	OA decomposition was studied by SRNL using manganese-catalyzed nitric acid.
1985	Tank 24 was cleaned using a similar regime as the Tank 16 regime with disappointing results. Tank 24 had large amounts of zeolite that is unaffected by OA.
1990s	SRNL and other laboratories completed several studies to understand the solubility behavior of actinides in acid solutions.
2000	A TFA-sponsored research effort explored Russian chemical cleaning methods. The Russians used a suite of dicarboxylic acids (obtained from an acid waste stream at a polyester plant) to dissolve the waste. The study concluded that the SRS OA regime was as effective and was safer than the alternative regime. However, the Russian researchers recommended striking aluminum-rich sludge first with concentrated caustic solution to leach those compounds, and to investigate augmenting OA with a safer acid such as citric.
2003	The Heel Removal Task Team was formed to investigate the best method to dissolve waste tank heels and to implement the method into the modern HLW program. The team performed a literature study of other chemicals and concluded OA to be the safest and most effective cleaning agent.
2004	Following up on studies conducted by Russian researchers, SRNL investigated sludge dissolution with citric and OA combinations.
2006	SRNL investigated nitric acid dissolution. Preliminary studies revealed high corrosion rates of waste tank material without substantial improvement of sludge dissolution.

Year	Development Description
2007	HLW engineering personnel investigated commercial nuclear plant cleaning methods for heel removal adaptation. Among others, the CORD-UV[1] process was proposed as viable. The process involves the decomposition of the oxalates as a central feature.
2007	The ECC project was formed to develop and deliver an OA cleaning process that would decompose the oxalates before entering the tank farms system.
2010	SRNL researched methods to increase chemical cleaning effectiveness of OA using supplemental acids.

1 CORD-UV - chemical-oxidation-reduction-decontamination with ultra-violet light destruction. A process used by a number of nuclear plant service providers to clean scale from stainless steel plant components.