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TANK CLOSURE CESIUM REMOVAL AT SAVANNAH RIVER SITE

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ABSTRACT

High-level waste (HLW) at the Savannah River Site (SRS) is currently stored in aging underground storage tanks. This waste is a complex mixture of insoluble solids (sludge) and soluble salts in an alkaline solution. Continued long-term storage of these radioactive wastes poses an environmental risk. The salt waste is currently treated to remove cesium by an interim solvent extraction process prior to disposal as grout in the Saltstone Disposal Facility. This interim process will be replaced in late 2018 with the Salt Waste Processing Facility (SWPF) to increase the salt waste treatment capacity; however, treatment and disposal of the salt waste is the critical path to waste retrieval, risk reduction and closure of old style tanks at SRS.

Savannah River Remediation (SRR) is developing a supplemental at-tank ion exchange capability for the removal of cesium from liquid salt waste. Building on the conceptual development of previous ion exchange initiatives and the experience of commercial nuclear plant decontamination services, and following the disaster response associated with Fukushima, the technology exists in industry to develop and deploy a modular at-tank treatment system for the selective removal of cesium from bulk salt waste effectively and efficiently. This Tank Closure Cesium Removal (TCCR) initiative would provide a supplemental treatment capability to existing and future salt processing, and improve confidence in supporting the desired acceleration of waste retrieval and tank closure efforts.

In addition to accelerating waste retrieval and tank closure activities, demonstration of this capability has several other benefits. Deployment of commercially available technologies with minor modification can expand Department of Energy (DOE) utilization of commercially supplied treatment processes. This “at-tank” treatment

system could be modified to evaluate and demonstrate removal of other radioactive constituents (strontium, technetium, iodine, etc.) of bulk waste. The modular aspect of this capability can allow for additional units to be delivered and deployed at SRS, as well as, provide supplemental treatment capabilities for waste treatment at other DOE sites.

BACKGROUND

HLW at SRS is generated from the chemical separations facilities as acidic raffinates. The raffinates are pH-adjusted prior to discharge to the tank farms to a pH of 14 for corrosion control of the carbon steel tanks. This waste exists either as a solid sludge phase, liquid supernate, and/or as crystallized salts. While stored in the waste tanks, the insoluble solids settle and accumulate on the bottom in the form of sludge. The remaining liquid volume is reduced by evaporating excess water. Continued evaporation of supernate results in concentrated salts crystallizing to form hard, but porous, salt cake.

Closure of liquid waste (LW) tanks is one of the major steps in the comprehensive remediation of the waste systems at SRS. The Federal Facilities Agreement (FFA) establishes the regulatory framework for the operation, new construction, and eventual closure of the LW tank systems. Non-compliant tanks (lacking full secondary containment) at the site are being closed in accordance with a formal agreement among the DOE, Region IV of the U.S. Environmental Protection Agency (EPA), and the South Carolina Department of Health and Environmental Control (SCDHEC) as expressed in the SRS FFA. Tank waste storage and removal are governed by an industrial wastewater operating permit issued by the SCDHEC. The FFA, Section IX.E., addresses the eventual removal of tanks and ancillary equipment from service and the final closure of the tanks.

A substantial majority of the waste volume stored at SRS is in the form of salt cake. As shown in Fig. 1, removal of salt waste requires re-introducing the water fraction to dissolve the salt and mobilize the waste.

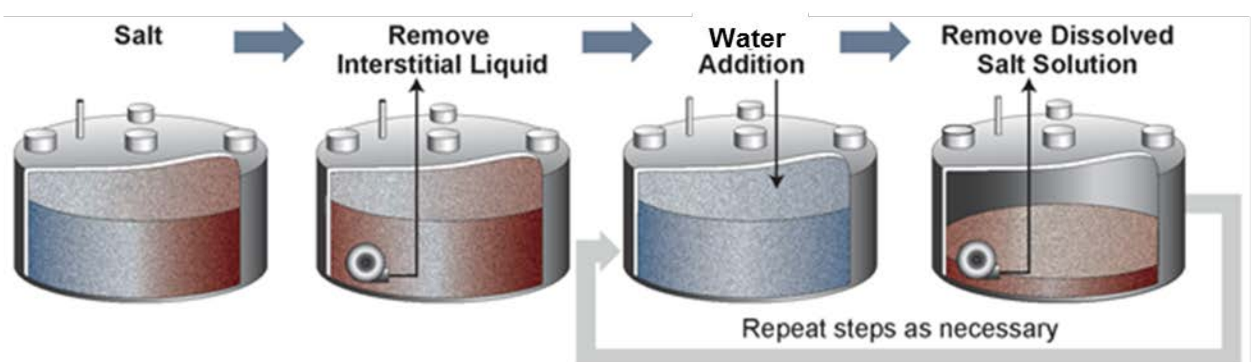

Fig. 1: Salt Removal Process

Dissolving the salt cake requires a significant amount of available tank space (approximately 3:1) to store the salt solution, and competes for available space with sludge waste retrievals, waste influents into the tank farm, and other ongoing treatment processes. Additionally, a treatment process must be readily available for processing the salt waste for disposition to alleviate tank space constraints and enable continued waste removal efforts.

PROCESS IDENTIFICATION

Previous SRS IX Pursuit

In an effort to reduce the activity level of evaporator effluent streams known as overheads, a polishing step using zeolite was developed to remove radioactive cesium. This was done by adding a zeolite packed column, known as the *cesium removal column* or CRC, to the treatment system.

The first column was installed in Tank 19F in November 1963 to treat the overheads from Evaporator 242-F. It was packed with Decalso™ ion exchange resin. Decalso™ is a man-made zeolite that was used experimentally at other DOE sites with good results. Eventually the Decalso™ resin was replaced by a Union Carbide manufactured resin called Linde AW-500 (eventually renamed IE-95). This is a natural (mined) zeolite made up of refine chabazite and erionite. Linde AW-500 proved effective in removing cesium from the overheads.

With improved vapor handling techniques employed in the evaporators, and upon completion and startup of the Effluent Treatment Facility (ETF), the use of CRCs was discontinued. The ETF now provides enhanced decontamination of the overheads stream prior to release to the environment making the utilization of the CRCs unnecessary.

More recently, ion exchange treatment processes have been proposed at SRS to supplement treatment (cesium and strontium removal) of salt waste and recover tank space lost as a result of technical issues and delays associated with SWPF. These initiatives focused on locating the majority of process equipment "in-tank" to leverage the containment and shielding capabilities associated with existing structures. Due to the costs associated with this larger scope and the lack of available tank space, these initiatives were not aggressively pursued.

Fukushima Experience

Following the Tohoku earthquake and subsequent tsunami which impacted Japan on the 11^{th} of March, 2011 a serious accident at the Fukushima Daiichi nuclear power plant resulted in a core meltdown and release of radioactivity. As a result of the accident an accumulation of thousands of gallons of highly radioactive sea water and some reactor coolant accumulated in a turbine building.

In response to the accident and the need to treat the radioactive water, units utilizing an ion exchange process were successfully deployed to treat the liquid with a cesium removal resin similar to the media that is being proposed for use at SRS.

Tank Closure Acceleration

Acceleration of tank closures is desired to better assure continued progress in accordance with the commitments among DOE, the EPA, various state agencies and community stakeholders. A readily deployable, supplemental cesium removal capability (ion exchange) has been determined to be advantageous in supporting this goal. This high impact challenge area will build on the experience of modular commercial nuclear plant decontamination following the disaster response associated with Fukushima. Various technologies exist in industry, and appear to have matured in capability and reliability, to accomplish larger scale, selective removal of the cesium component of the bulk salt waste effectively and efficiently. Successful completion of this high impact challenge will demonstrate deployability of low cost, modularized, targeted treatment capabilities as DOE's solution of choice and serves as another example that commercial industry can develop and provide capable solutions to help solve DOE's complex-wide challenges.

PROCESS DESCRIPTION

The TCCR initiative consists of an ion exchange process for the removal of cesium from liquid salt waste to provide a supplemental treatment capability and improved confidence in supporting the desired acceleration of waste retrieval and tank closure efforts. This at-tank treatment system would be developed by a commercial supplier and delivered to SRS for installation and conducting a full scale demonstration of the capability to treat waste in Tank 10.

A Statement of Work (SOW) was developed in 2015 to solicit proposals from industry to develop and deliver the TCCR system [1]. A commercial supplier will design, fabricate, test and deliver an "at-tank" modular cesium removal system to be deployed at SRS for the treatment of liquid salt waste from Tank 10H. The SOW provided expected waste characterization information, defined the general scope of the TCCR system and established minimum performance criteria for TCCR system development by the Supplier. SRR will install the TCCR system in the SRS tank farm adjacent to Tank 10H. Additionally, SRR will design and implement systems to dissolve salt and deliver the salt solution to TCCR, receive and disposition the treated salt solution from TCCR, and the interfacing systems necessary to support TCCR operation. A simplified diagram of the TCCR system is shown in Fig. 2.

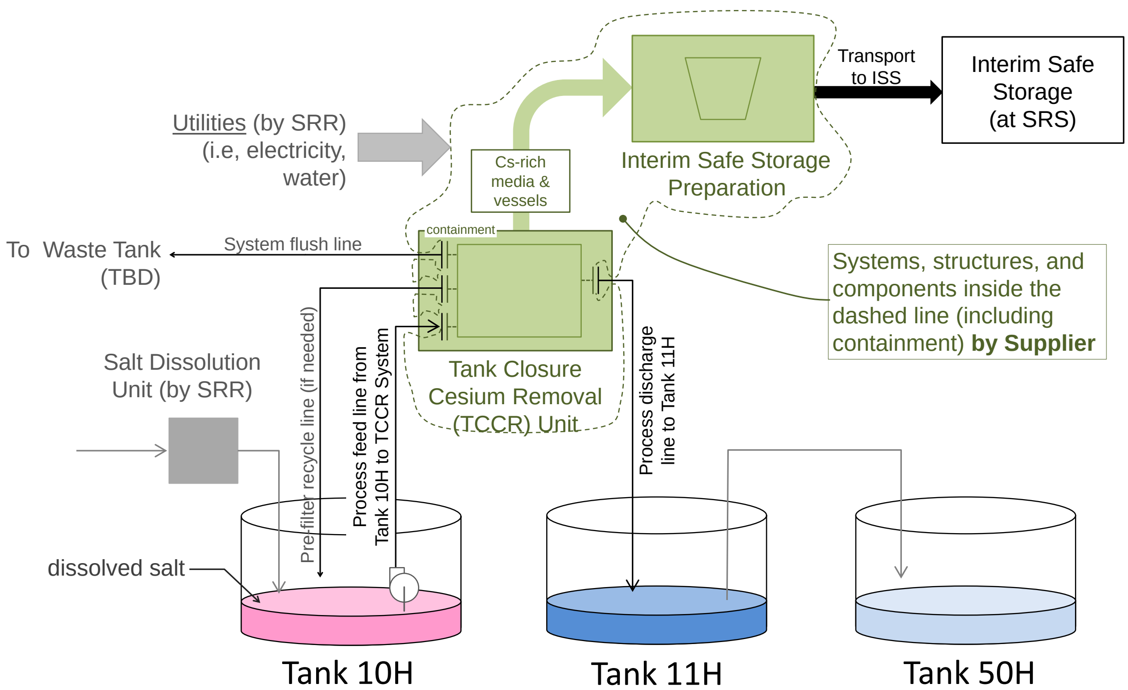

Fig. 2: TCCR System Simplified Diagram

Salt waste would flow through a pre-filter to reject any solid material from entering the treatment system and be returned to Tank 10H. Clarified salt waste would then flow from the pre-filter into the first stage ion exchange column. The ion exchange media utilized will be highly selective for the removal of cesium and allow most other soluble constituents to pass through the media. Additional ion exchange columns may be provided in series to improve decontamination factor, protect against flow "channeling" in the column, guard against media breakthrough, and allow for more continuous operation of the system. Decontaminated (cesium removed) effluent would flow from the ion exchange columns and sent to Tank 11H prior to being transferred to Saltstone for disposal.

Once the ion exchange media in a column becomes loaded with cesium to the extent practical (spent), the column with media will be removed from the system and replaced with a new ion exchange column loaded with fresh media. The spent column will be temporarily stored in an Interim Safe Storage (ISS) location within the SRS Tank Farm Facility. This ISS concept reduces initial need for back-end process facilities, defers the cost of these facilities, and allows for identification and evaluation of potential future disposal alternatives.

Performance capabilities of the TCCR system were established to be comparable with existing salt treatment processes and treat the volume of salt waste in Tank 10H in a reasonable amount of time. The remaining waste in Tank 10H contains approximately 3700 Terabecquerel (TBq) (100,000 curies (Ci)) of cesium-137. The minimum cesium removal efficiency, or decontamination factor (DF), is 1000. Dissolution of the salt cake will be performed in a batch fashion of approximately 6 batches, with a total expected volume of ~2.27 E+6 liters (600,000 gallons). A minimum flow rate through the unit was established at 19 liters per minute (5 gpm) in order to treat Tank 10H in less than 6 months. TCCR performance attributes and the desired feature are summarized in TABLE I.

TABLE I: TCCR System Performance Attributes and Benefits

Attribute	**Benefit**
Engineered mobile enclosure	Provides treatment portability
Modular design	Major components removable
Pre-filtration	Reduce insoluble solids and prolong bed life
5 gpm minimum processing rate	Rate selected to balance size, cost, schedule
Decontamination factor ≥ 1000	DF necessary to not impact disposal
Multiple IX columns	Flexibility for continuous processing
Column design	Balance size, shielding, heat load
Integral column shielding	Reduces column rad rates to 5 mrem @ 30 cm
Post-IX filtration	Retains resin, fines within system
ISS for 10 years	Allows development of alternatives and implementation of disposition pathway

BENEFITS

Modular Treatment System

At-tank Treatment Capability

The TCCR unit is being designed as a modular arrangement. The enclosed, shielded process will be mounted on skids that can be deployed to different locations throughout the tank farms. This design attribute is crucial in the ability of the TCCR system to accelerate tank closure not just at SRS, but throughout the DOE complex.

Modular Design

One of the key design features that will allow the TCCR process to be utilized in the tank farms and other DOE sites is the ability to remotely remove and replace the ion exchange columns. When a column is loaded to the desired capacity, it can be removed and replaced with a new column that is ready for processing. The removable columns allow for the processing of waste that can vary from tank to tank. For example, if the Cs-137 concentration varied from tank to tank, the total number of columns used to treat that specific waste stream (equivalent bed length) could be varied to control cesium removal efficiency, or the individual column design could be modified to maximize media efficiency for a given concentration. This design offers the TCCR system a great amount of flexibility in its treatment capabilities.

Radioactive Test Bed

Full-scale CST Demonstration in Alkaline Waste

While the CST ion exchange media has been used in previous cesium removal applications, the media was not subjected to the same treatment conditions as SRS salt waste. The TCCR system, which will be ready for demonstration in 2018, will use four ion exchange columns to remove approximately 11,100 TBq (70,000 Ci) of cesium from Tank 10H salt solution. Each column will be loaded to a capacity of approximately 3700 TBq (25,000 Ci) per column.

Previous CST demonstrations have used similar waste streams as feed to the ion exchange system. For example, during the 1997 demonstration at Oak Ridge National Lab (ORNL), actual waste from the Melton Valley Storage Tanks (MVST) was processed through smaller ion exchange columns filled with CST. The waste from the MVST was similar to the SRS waste; however, the concentration of Cs-137 in the MVST feed was one order of magnitude less than the expected concentration from Tank 10H. The maximum column loading seen during this demonstration was about one order of magnitude below the design requirements for the TCCR system. The ability of the TCCR system to load the CST with a higher concentration of Cs-137 shows a more efficient use of ion exchange media, which could save the DOE complex valuable resources as well as reduce the final volume of spent ion exchange material.

Targeted Radionuclide Removal

Although cesium removal is the initial focus, various radionuclides exist in wastes that are beneficial targets for removal. One or more of the columns in the TCCR system could be designed and loaded with other ion exchange materials that are designed to remove the other radionuclides (eg., strontium, technetium, etc.). Although the initial system is designed for inorganic media, organic resins such as SuperLig® 639 could be demonstrated with TCCR for various waste streams. Additionally, columns can be filled with multiple ion exchange materials in order to create a mixed bed system that would be customized to a given feed stream.

DOE Complex Benefits

Ion exchanger development continues to advance; new media and elutable resins continue to be developed. Having the capabilities to remove and replace columns, would allow cutting-edge technologies to be tested in a full-scale, practical application. For instance, the elutable resin, spherical resorcinol-formaldehyde, could be used in a scaled demonstration of cesium removal for Hanford.

Treatment of Other Waste Streams

The ability to customize the treatment capabilities of the TCCR ion exchange system lends itself to the treatment of wastes other than just dissolved salt solution. Recycle waste from the Defense Waste Processing Facility (DWPF) eventually must have an alternate treatment method. Tank heel remnants may not be conducive to solvent extraction. These streams are potential candidates for future feed to an ion exchange system. The TCCR unit could be tailored to demonstrate waste treatment from other DOE sites as well.

Alternative Disposition Pathways

The columns are designed with integral shielding for complete removal from the system for storage. Interim safe storage (ISS) of spent media and columns for a maximum of ten years allows for evaluation and selection of the most suitable disposal pathway.

The CST media was selected due to its high selectivity for cesium, low volume required, radiation and thermal stability, solid cesium end form, and no radiolytic hydrogen generation when the resin is dry. Columns are designed and will demonstrate the capability for media removal by sluicing. Columns also have the capability for direct disposal in a radioactive landfill. The high selectivity for cesium, a gamma emitter with a half-life of only 30 years, provides the opportunity to re-characterization the spent media for disposal as low level waste.

CURRENT STATUS

SRR awarded a fixed price subcontract for the design, fabrication, testing and delivery of the TCCR process equipment to Westinghouse Electric Company (known as the TCCR subcontractor) on July 7, 2016, with a 15 month period of performance. The TCCR equipment will utilize a modular design concept to accommodate minimal onsite assembly and preparation work. SRR will site the TCCR equipment in the vicinity of Tanks 10 and 11 such that the TCCR equipment will be sufficiently compact to minimize the processing area footprint, the length and number of above-grade transfer lines, and the volume of liquid radioactive material outside of the tank.

Nuclear safety analysis and hazards assessment of TCCR will be conducted by SRR with participation from the TCCR Subcontractor. SRR will install the TCCR equipment at the design location, provide Balance of Plant (BOP) activities (site preparation, utilities, services, transfer lines and tie-ins), startup the process, develop procedures and operator training and complete acceptance and commissioning activities. SRR will perform the TCCR demonstration operation to treat the dissolved salt solution from Tank 10 and will perform a post demonstration evaluation on TCCR effectiveness following completion of demonstration operation.

SRR maintains a Responsibility Matrix with the TCCR Subcontractor and is shown in Fig. 3.

Activity	Supplier	SRR
Design, fabricate, and deliver the TCCR System	✓	
Submit documentation for the TCCR System	✓	
Conduct nuclear safety analysis	◊	✓
Conduct hazards analysis	◊	✓
Assign safety classification to SSCs		✓
Conduct factory acceptance testing of TCCR System per SRR-accepted procedures (includes purchasing, preparation and disposition of simulant)	✓	
Install TCCR System at Tank 10H and 11H	◊	✓
Perform site acceptance testing of TCCR System	◊	✓
Conduct training of SRR operations personnel	◊	✓

✓ Responsible (lead)

◊ Support role only (e.g., technical assistance – no hands-on work)

Fig. 3: TCCR Subcontractor Responsibility Matrix

As of November 2016, all ongoing TCCR activities are on schedule as shown below:

- The TCCR Subcontractor achieved the first three design contract milestones, including the preliminary and intermediate design deliverables for the TCCR equipment. The TCCR Subcontractor is on schedule to complete within the 15 month period of performance.
- The activities for the nuclear safety in design consolidated hazards assessment have started and are on schedule.
- SRR Balance of Plant and utility design activities have started as planned

Prior to shipment in September 2017, the TCCR Subcontractor will complete a Factory Acceptance Test (FAT) of the TCCR equipment utilizing a salt waste simulant to conduct the test. The test requirements are included in the TCCR subcontract Statement of Work. Attributes of the TCCR Supplier FAT will be utilized for the Readiness Assessment. The TCCR FAT will include tests for:

- Pre-Startup: System testing, instrument calibration, continuity checks, alignment, etc.
- Cold Run: Column and filter performance, hydraulic flow testing, ventilation, etc.

- Ion Exchange Column Testing: Column differential pressure and absence of channeling
- Maintenance: Column dewatering, column and filter removal, resin removal, etc.
- Storage System: Demonstrate handling and storage

The TCCR Demonstration will be performed in Tank 10 and is planned for 6 batches of approximately 3.785 E+5 liters (100,000 gallons) each, with a processing throughput of a nominal 19 liters per minute (5 gpm), for a total duration of 4-5 months. The decontaminated salt solution will be transferred to Tank 11. The TCCR Demonstration of operations in planned for completion in August 2018.

A post demonstration evaluation of TCCR performance will be documented to provide a feasibility analysis to DOE-SR regarding use of the commercially-sourced technology. The post demonstration evaluation will be complete in December 2018. A summary level schedule for development and deployment of TCCR is shown in Fig. 4.

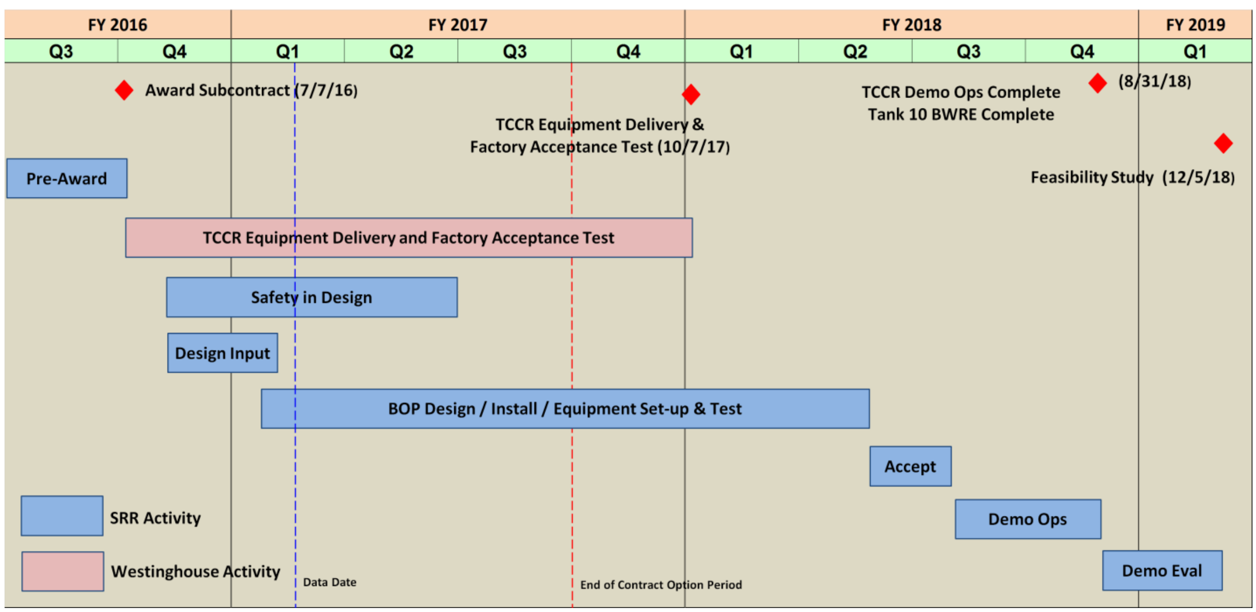

Fig. 4: TCCR Summary Schedule

SUMMARY

A supplemental cesium removal capability utilizing ion exchange technology is advantageous in supporting accelerated waste retrieval and tank closures. The TCCR system leverages modular treatment capabilities demonstrated during treatment of sea water at Fukushima. A commercial supplier is designing and fabricating the system for technology demonstration with Tank 10H waste.

This system advances technology development of the following:

- Modular design to enable:
  - Mobile, at-tank treatment capability
  - System modularity to adapt to varying feed streams
- Radioactive test bed for:
  - Full scale CST demonstration with SRS alkaline waste
  - Other targeted radionuclide removal with selective ion exchange media or mixed beds
  - Potential for treatment demonstration for other DOE sites
  - Supplemental treatment of other waste streams (e.g. DWPF recycle)
- Development of alternative disposition pathways

REFERENCES

1. Tank Closure Cesium Removal System Statement of Work, X-SOW-H-00002 Revision 3, January 4, 2016