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Summary of Batch Operations for the Tank Closure Cesium Removal Continued Operations (TCCR 1A) at SRS

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ABSTRACT

Savannah River Remediation (SRR) manages and operates the liquid waste facilities at Savannah River Site (SRS) for the Department of Energy (DOE). Stored liquid waste is a complex mixture of insoluble solids (sludge) and soluble salts in an alkaline solution. SRR has deployed the Tank Closure Cesium Removal (TCCR) system, a tank-side ion exchange process, to remove radioactive cesium from salt waste and enable onsite disposal of the resulting decontaminated salt solution as low-level waste at the Saltstone facilities. The TCCR system consists of two prefilters, four ion exchange (IX) columns, one resin trap, and a ventilation system. The IX process uses a form of inorganic crystalline silicotitanate (CST), which has a high affinity for cesium and other alkali metals, strontium, and actinides. The TCCR Demonstration Project has processed approximately 1.06 million liters (280,000 gallons) of salt feed from Tank 10. Approval has been received from DOE to continue TCCR operations processing Tank 9 material under the project name TCCR 1A.

The feed for TCCR 1A is created from saltcake in Tank 9 through a dissolution process. Once enough salt has been dissolved, this salt solution is transferred to Tank 10 and qualified. The qualification process characterizes the feed which may need chemical adjustment to ensure that the cesium loading on the columns will not cause boiling of waste within the columns during or after processing. Once the batch has been qualified, salt waste is fed to the TCCR system through a submersible transfer pump in the Center Riser of Tank 10. The waste is filtered through a set of two parallel shielded, dead-end prefilters that prevent solids buildup in the columns. The filtered salt solution then travels to the shielded IX columns, which can be operated individually or in series, where the cesium is sorbed onto the CST media. The decontaminated salt solution (DSS) then proceeds through a resin trap and out of the module to Tank 11. After the IX columns are spent, they are removed from the process enclosure and relocated to the Interim Safe Storage Facility (ISS) to await eventual disposition.

TCCR 1A has successively processed approximately 320,000 liters (85,000 gallons) of salt waste during its first batch. Processing of this batch will serve as a further input to the TCCR technology demonstration report. This paper provides an operational summary of the first TCCR 1A batches, and how it will be used as an input to the TCCR technology demonstration report in conjunction with the three batches from the TCCR Demonstration Project.

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INTRODUCTION

Savannah River Remediation LLC safely manages and operates the liquid waste facilities at the Savannah River Site (SRS) for the Department of Energy (DOE). The SRS liquid waste system consists of 51 waste storage tanks, waste evaporators, treatment facilities, Defense Waste Processing Facility (DWPF) vitrification plant, and Saltstone Production Facility (SPF) grout solidification plant. Waste processed through the latter is being disposed of onsite in Saltstone Disposal Units and DWPF vitrified waste is destined for a federal repository. Eight of the 51 waste storage tanks have been operationally closed and filled with grout. As liquid waste treatment and disposal continues, the remaining storage tanks and ancillary infrastructure will be closed in the coming years.

Radioactive waste at SRS was generated from the chemical separations facilities. The waste is pH-adjusted to a high pH for corrosion control of the carbon steel tanks prior to transfer to the tank farms for storage. During storage in the waste tanks, 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 crystallized concentrated saltcake.

A treatment process to remove cesium, strontium, and soluble actinides from the salt waste is required to enable continued waste removal efforts. The salt waste was previously treated by an interim filtration and solvent extraction process, Actinide Removal Process/Modular Caustic-side Solvent Extraction Unit (ARP/MCU), prior to disposal as grout in the Saltstone Disposal Facility. This interim filtration and solvent extraction process has been replaced with the higher capacity Salt Waste Processing Facility (SWPF). Even with increased salt treatment capacity, treatment and disposal of the salt waste remains critical to risk reduction, waste retrieval, and closure of old-style tanks at SRS.

The TCCR System is a modular filtration and ion exchange (IX) process for the removal of cesium from liquid salt waste. This at-tank system was initially designed to process up to 2.37 ML (625,000 gal) of salt waste from Tank 10H, removing Cs-137 to meet design criteria and downstream processing constraints [1]. To meet production goals, the TCCR system needs to process the salt from both Tanks 9H and 10H while removing enough Cs-137 and other radionuclides, through either ion exchange or filtration, to meet the Waste Acceptance Criteria (WAC) of SPF where the salt will be permanently dispositioned.

PROCESS DESCRIPTION

The TCCR Demonstration Project is an at-tank modular filtration and IX process currently deployed on Tank 10H. Prior to entering the TCCR system, the salt feed batches are prepared by dissolving salt in Tank 9H by adding water prior to transfer to Tank 10H. After any necessary addition of chemical adjustments as determined by process chemistry, the salt solution contents are recirculated until a constant density is achieved. After recirculation of the salt solution is stopped, the batch qualification process begins.

Qualification of the batch is performed using both a batch equilibrium contact test (BECT) and sample results to meet several qualification criteria. A BECT is performed in Tank 10H by contacting approximately 0.1 g CST solids with the large volume of Tank 10H radioactive waste for a minimum of 10 days [2]. These samples are then analyzed for cesium content. The feed meets one of the qualification criteria if the cesium content is below allowed limits established in the safety basis for TCCR operation. Other qualification values set to protect safety basis values include a minimum nitrite concentration, minimum feed batch temperature limit, and limits on IXC backflushes and liquid additions to Tank 10H. Sample results are used to confirm the nitrite concentration is above 0.0275 M to inhibit corrosion, as well as catalytic and radiolytic hydrogen generation effects from organic compound contributions. A minimum feed batch temperature limit is set to protect the BECT results as lower temperatures favor cesium sorption by the CST. The qualification of a TCCR batch also sets limits on total number of IXC backflushes and total volume of liquid additions to Tank 10H without invalidating the qualification. Backflushes of a used IXC may contain cesium that has desorbed from higher temperatures within the column and increases the cesium concentration in Tank 10H. Liquid additions to Tank 10H may dilute the supernate and cause more salt to dissolve after conclusion of qualification efforts. These operational evolutions are limited to protect the loading of Cs-137 on the IXC assumed in the safety basis and verified by the BECT. [3]

Upon qualification of the salt batch, radioactive salt solution flows from Tank 10H through hose-in-hose (HIH) transfer lines to the TCCR unit. The TCCR unit consists of three primary components: a main process enclosure containing all components necessary to treat the salt waste, a ventilation skid to provide negative pressure for the main process enclosure, and a control room to house the operating control system, operator interface, and video monitoring systems. Once the salt waste enters the main process enclosure through the HIH transfer line, the salt waste flows through a prefilter where any solids are removed from the stream and returned back to Tank 10H. The filtrate flows into one or more ion-exchange columns (IXCs) containing a cesium-specific crystalline silicotitanate (CST) media, UOP IONSIV R-9120B 30x60 non-CW. Prior to installation within the TCCR Unit, the IXCs underwent fines removal after being filled with CST and have been treated with 3 M sodium hydroxide to convert the CST to the sodium form. Cesium, minor amounts of Sr-90, and some soluble actinides are removed from the salt solution via contact with the CST media. The resulting decontaminated salt solution (DSS) exits the IXCs and passes through a nominal 80 μm post-filter, or resin trap, which captures any solids that may be in the column effluent prior to exiting the TCCR unit to Tank 11H. A simplified process diagram of the TCCR system is shown in Figure 1. [4]

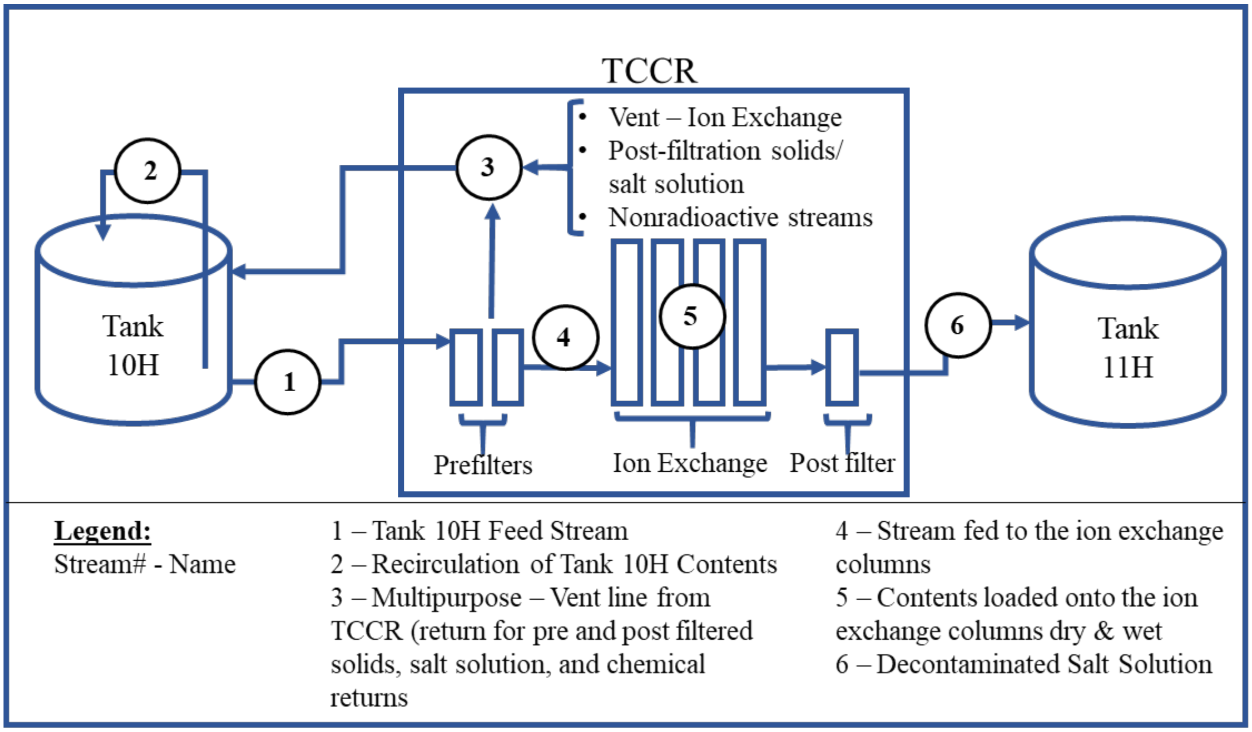

Fig. 1. Major Radionuclide Streams for TCCR.

This system was designed to treat 2.37 ML (625,000 gal) of Tank 10H radioactive salt solution. The TCCR system started radioactive operations in 2019 and has successfully processed ~1.14 ML (~300,000 gal) of salt solution from Tank 10H across three batches.

PROCESSING RESULTS

Batch Preparation & Characterization

Table I summarizes the major results for the preparation and characterization of TCCR 1A Batch 1. Specific parameters for each batch are discussed in the subsequent sections.

TABLE I. Summary of TCCR 1A Batch 1 from Tank 10H

	Parameter	**Batch 1***
Batch Preparation	Total Additions to Tank 10H (L and [gal])	TBD
	Batch Size Processed through TCCR (L and [gal])	
Supernate Characteristics	Density [g/mL]	
	Sodium (M)	

	Carbonate (M)	
	Sulfate (M)	

Batch 1

TBD

Prefiltration Performance

The TCCR unit contains two dead-end prefilters arranged in parallel. The previous models of filters consisted of 19 tubes approximately 35.6 cm (14 in) long and 2.5 cm (1 in) in diameter with an absolute filtration rating of 10 µm. One prefilter is online during processing. Once the differential pressure (dP) across the online prefilter reaches a predetermined setpoint, a backwash is initiated. The feed is aligned to the other prefilter and filtrate is redirected to the backside of the filter that needs to be backwashed. The flowrate during a backflush sequence is approximately 1.5 times that of the target flowrate to the IX columns. Once the backwash duration has been reached, filtrate is aligned to the IXCs and normal flow is re-established with the other filter online. The backwash duration is a predetermined value that can be changed during processing. Performance data of interest for the prefilters is differential pressure (dP) across the filter, time between filter swaps from the beginning to the end of the batch, and recovery of flow and differential pressure following a backwash. These parameters can be seen in Table II for each of the batches.

Table II. TCCR Pre-filter Performance for Batch 1

Parameter	Batch 1A
Baseline dP (kPa and [PSI])	TBD
dP Setpoint for Backflush (kPA and [PSI])	TBD
Observed Pre-filter Swap Frequency (hours)	TBD
Target Flowrate to IX Columns (L/min and [gpm])	TBD
Back Wash Time (seconds)	TBD

Batch 1

TBD

IXC Hydraulic Performance

Pressure gauges monitor the pressure across all online IXCs. These readings are recorded by the control system 2 times per second. During the design of the TCCR System, a model was created to predict the hydraulic performance of the columns. It required a user input of number of IXCs online, flowrate, and specific gravity, with assumed pressure drops across the IXC from historical data. The model predicted a differential pressure (dP) across 2 IXCs at a flow rate of 19 L/min (5 gpm) of approximately 91.4 kPa (13.25 psi) and 63.4 kPa (9.2 psi) when the feed has a specific gravity of 1.5 and 1, respectively. At a flow rate of 38 L/min (10 gpm), the predicted dP increases to 235 kPa (34.09 psi) and 160 kPa (23.2 psi) when the feed has a specific gravity of 1.5 and 1, respectively. The model also predicted dP across 4 IXCs at a flow rate of 19 L/min (5 gpm) of approximately 165 kPa (24 psi) and 113 kPa (16.4 psi) at a feed specific gravity of 1.5 and 1, respectively. The dPs at a given flowrate and IXC configuration have some variation due to the flow control valve and backflush frequency during processing of the salt batch. Pressure across the IXCs equalize during a prefilter backflush sequence and shows a negative value due to the difference in height between the gauges.

The three TCCR Demonstration batches operated with varying flowrates and IXC configurations. The flowrates, number of online IXCs, and resulting average dP across the online IXCs are shown in Table III.

TABLE III. TCCR IXC Hydraulic Performance for Batches 1

Parameter	Batch 1
Flowrates Tested (L/min and [gpm])	TBD
1-IXC dP (average kPa and [average psi])	
2-IXC dP (average kPa and [average psi])	
3-IXC dP (average kPa and [average psi])	
4-IXC dP (average kPa and [average psi])	

Batch 1

TBD

Ion Exchange Column Loading

The TCCR unit IXC cesium loading performance may be measured by comparing Cs-137 activity in the feed stream with activity in the column effluent. Currently, there are two methods

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available to perform this comparison for the TCCR unit. The first is comparing the supernate samples from Tank 10H and Tank 11H and the second is comparing the field readings of the radiation monitors for the feed stream and the exiting column effluent.

Tank 11H is the receipt tank for the DSS. It was previously a sludge slurry tank and has completed sludge removal activities. Despite the best efforts of sludge removal, some sludge mounds remain in the tank. Water rinses were performed to lower the radioactivity caused by soluble radionuclides leaching from the sludge. The supernate volume in Tank 11H was reduced to a minimum before operation of TCCR, so it is expected that the final composition in Tank 11H after processing would resemble the TCCR feed concentrations with the main exception of Cs-137. However, the water rinses did not eliminate leaching of soluble radionuclides into the more dilute DSS solution as it sits in contact with the sludge heel during processing of the batch through the TCCR unit. Quantification of this leaching behavior has not yet been performed and the impacts to the DSS from the heel in Tank 11H have not been discerned.

Radiation monitors were used throughout processing to monitor key steps of the TCCR process. The monitors read the gamma emission down to levels as low as 0.0001 mRem/hr from Ba-137m, which is normally in secular equilibrium with Cs-137. The minimum value reported by the operating system is 0.1 mRem/hr, even if the monitor indicates a lower value. Therefore, all decontamination factors (DF) reported by the TCCR operating system from these values would be biased significantly low. However, the CST used in the IXCs is also highly selective for Ba-137m and removes most of the Ba-137m from the feed. The DSS monitor is not positioned to permit the Cs-137/Ba-137m ratio to return to secular equilibrium, but rather at a position where only approximately 1 half-life has passed, at a flow rate through TCCR of 19 L/min (5 gpm). Consequently, the gamma measurement by the radiation monitors reflects only about one half of the approximate Cs-137 concentration.

Efforts were made to improve the ability to understand the performance of the unit by improving detection capability for TCCR 1A Batch 1. These efforts included the design and installation of new radiation monitors that have a more sensitive detection limit and can account for varying process flow rates, as well as a modification to the discharge line that maintains a full pipe volume at the detection area. The varying flow rate changes the number of Cs-137 to Ba-137m half-lives that have elapsed between the time DSS leaves the column and passes the monitor. They also included more frequent sampling from Tank 11H to reduce the time exposed to the Tank 11H heel and more accurately characterize the Cs-137 removal of the IXCs. The more frequent sampling showed a decrease in Tank 11H cesium concentration as processing progressed, indicating cesium removal by the columns.

Despite the difficulty in obtaining an accurate value, it is estimated based on the totality of information available, that the TCCR unit achieved the minimum design requirement bulk average decontamination factor (DF). Additionally, TCCR 1A Batch 1 has met the SPF WAC for permanent disposition.

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CONCLUSIONS

The TCCR Demonstration project successfully processed 1.14 ML (300,000 gal) of radioactive salt solution from Tank 10H and the continued operations of TCCR 1A has processed an additional approximately 320,000 liters (85,000 gallons).

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