Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-09SR22505 with the U.S. Department of Energy (DOE) National Nuclear Security Administration (NA).

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TCCR Operational Summary and Optimization for Tank 9 Processing - 20314

R.A. Seeley, T.L. Fellinger, A.I. Silker, A.M. Luzzatti, R.O. Voegtlen & G.C. Arthur
Savannah River Remediation LLC, Aiken, SC 29808

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. This process is currently deployed utilizing salt feed from Tank 10, with future plans to dissolve solid salt in Tank 9 and transfer the salt solution to Tank 10 for processing through TCCR.

The feed for TCCR must be created from saltcake in Tank 10 through a dissolution process. Once enough salt has been dissolved, a qualification process is entered. This process characterizes the feed and ensures 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 transfer pump in the center of the tank. The waste is filtered through a set of two 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 on the CST media. The decontaminated salt solution (DSS) then travels through a resin trap and out of the module to Tank 11.

TCCR has successfully processed approximately 795,000 L of Tank 10H radioactive salt waste over two batches to date. There has not yet been a system induced shutdown. The prefilters performed as expected with only minor degradation in recovery of differential pressure after a backflush sequence. The time between backflushes decreased as each batch reached the end of processing. The hydraulics in the IXCs mostly performed as expected at all flow rates, except for one IXC that will be further investigated during Batch 3 processing.

The TCCR system has shown some opportunities for more efficient processing during the length of the demonstration so far. For future processing of material from Tank 9H through Tank 10H and the TCCR unit, TCCR 1A will implement changes to the prefilters and the IXCs. The prefilters will have an increased surface area and a new filter media in an effort to increase time between filter swaps and improve backwashing cleaning capability. The IXCs will have a reduced diameter to allow for increased heat transfer out of the column and increased loading of Cs-137. Additionally, a new form of CST with an increased kinetic performance is being investigated for use during TCCR 1A operation.

INTRODUCTION

The Savannah River Site (SRS) Liquid Waste System safely stores and treats high-level radioactive waste. The Liquid Waste System consists of 51 waste storage tanks (eight of which are filled with grout and operationally closed), waste evaporators, treatment facilities, and solidification facilities such as the Defense Waste Processing Facility (DWPF) and Saltstone Production Facility (SPF). Radioactive waste at SRS was generated from the chemical separations facilities. The waste is pH-adjusted to a pH of 14 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 salt cake.

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 is currently treated by an interim filtration and solvent extraction process prior to disposal as grout in the Saltstone Disposal Facility. This interim filtration and solvent extraction process will be 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 approximately 100,000 Ci of Cs-137 with a minimum decontamination factor (DF) of 1000. [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 adding water to Tank 10 salt cake. After the addition of water, the salt solution contents are recirculated until a constant density is achieved. Recirculation of the salt solution is stopped upon a constant density reading and batch qualification begins.

Qualification of the batch is performed using both a batch equilibrium contact test (BECT) and sample results. A BECT is performed in Tank 10H by contacting approximately 0.1g CST solids with the large volume of Tank 10H radioactive waste for a minimum of 10 days [Ref. 2]. These samples are then analyzed for cesium content. The feed meets the 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, catalytic, and radiolytic effects on organic compound

contributions to hydrogen generation. A minimum feed batch temperature limit is set to protect the BECT results because lower temperatures favor cesium adsorption by the CST. The qualification of a TCCR batch also sets limits on total number of IXC backflushes and total volume of liquid additions 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. [Ref. 3]

Upon qualification of the salt batch, radioactive salt solution flows from Tank 10 through hose-in-hose (HIH) transfer lines to the TCCR unit. The TCCR unit consists of three skids: 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 skid 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 R-9120B. The columns underwent fines removal after installation 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 post-filter, or resin trap, which captures any resin that may be in the column effluent prior to exiting the TCCR unit to Tank 11. A simplified process diagram of the TCCR system is shown in Figure 1. [Ref. 4]

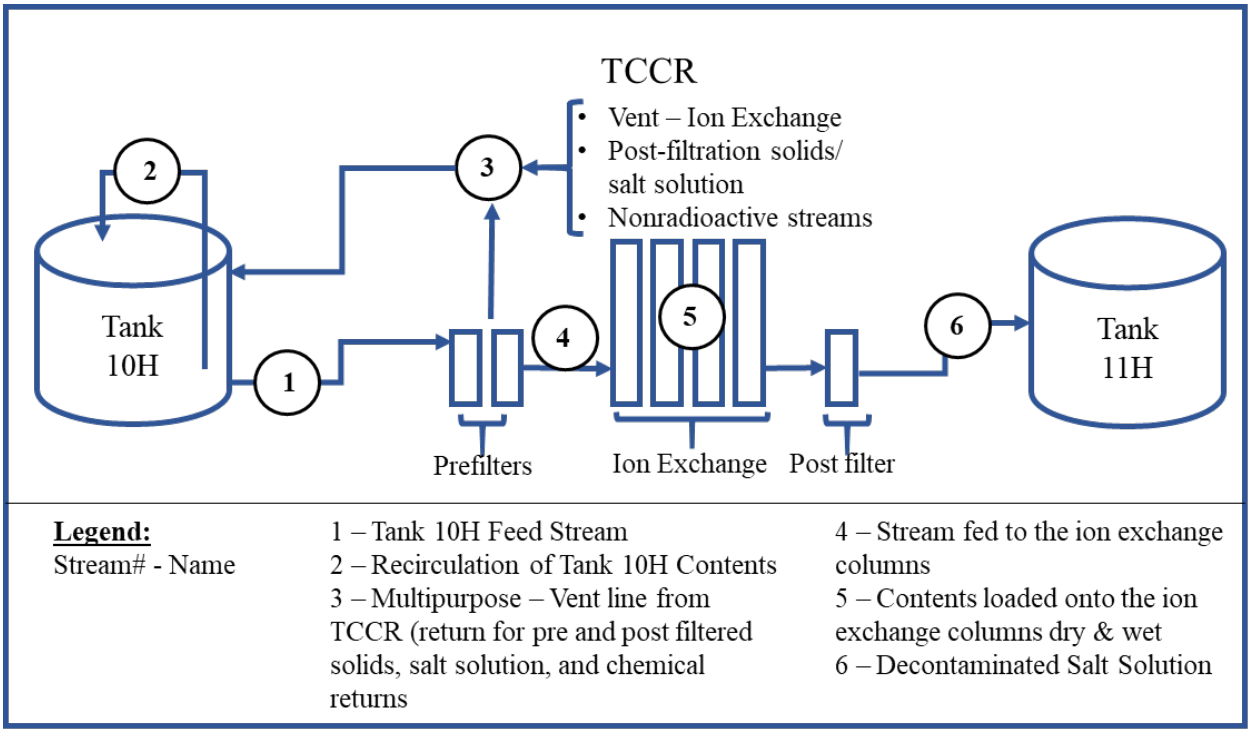

Fig. 1. Major Radionuclide Streams for TCCR.

This system was designed to treat 2.37 ML (625,000 gal) of Tank 10H radioactive salt solution. To date, TCCR has operated two batches. Approximately 575,000 L (152,000 gal) were processed during the first batch between January and February 2019. On June 21, 2019, TCCR began processing Batch 2. Approximately 220,000 L (58,000 gal) of salt solution from Tank 10 was processed successfully. Batch 2 processing terminated on June 29, 2019.

PROCESSING RESULTS

Batch Preparation & Characterization

Batch 1 and Batch 1A

TCCR Batch 1 was prepared in November 2018 by adding approximately 600,000 L (160,000 gal) of well water to Tank 10H. Recirculation in the tank was performed for seven days before density readings from a bubbler assembly on the pump indicated the target density between 1.2 and 1.3 g/mL. The qualification process began by adding two BECT assemblies to the tank and pulling duplicate surface samples of the supernate. The results of the surface samples indicated a lower than expected density of 1.089 g/mL and a sodium molarity of 2.02 M. The sodium (Na) molarity was lower than required (3.22 M) and required remediation of the batch with the addition of ~64,300 L (17,000 gal) of 50 wt% sodium hydroxide to meet a target concentration of 3.5 M Na.

After remediation of the batch to meet the minimum Na requirement, the batch was renamed Batch 1A. A density probe was deployed into Tank 10H at varying heights to indicate if the supernate was well-mixed. A second set of surface samples were obtained and a second set of BECT assemblies were deployed. The results of these surface samples, shown in Table I1, indicated a density of 1.164 g/mL, matching those of the density probe, and a sodium concentration of 3.79 M. The CST in the BECT assemblies was analyzed for loading results. They indicated a cesium loading of 8.96×10^{-3} mmol/g_{CST} or 23.2 $Ci_{Cs\text{-}137}/kg_{CST}$. This value was slightly lower than the expected value based on equilibrium modeling of Tank 10H composition. This may be partly explained by the loading of non-cesium constituents like calcium and iron that loaded to a greater extent than cesium (See Tables II and III for competitor loading).

These sample results were compared to qualification values and determined that Batch 1A was qualified to feed to the TCCR unit.

Batch 2

Changes were made to the preparation of TCCR Batch 2 in an effort to increase loading on the CST and more efficiently dissolve salt. Calcium was shown to load on the CST in the Batch 1A BECT. Potential sources of calcium include the saltcake and the well water. Domestic water was added to Tank 10H instead of well water in an effort to decrease the calcium in the supernate for Batch 2. Poor salt dissolution in Batch 1A sparked an evaluation of water additions for Batch 2. The poor dissolution was determined to be from a layer of difficult to dissolve double carbonate and sulfate salts in the salt cake. An equilibrium model was developed to take this into account and determine the optimal batch size for dissolution while maintaining the Na concentration above 3.22 M. Modeling results indicated two additions of 95,000 L (25,000 gal) with mixing in between additions would result in optimal dissolution.

Batch 2 was prepared with two 95,000 liter (25,000 gal) additions of domestic water. Seven days of recirculation was performed between the additions. Recirculation continued for approximately 2 months, at which time the density probe indicated consistent density readings at various heights throughout Tank 10H that had stopped increasing. Two BECT assemblies were deployed for the 10 day test and two surface samples were taken for analysis. The results , shown in Table II, indicated a density of 1.161 g/mL and a sodium concentration of 3.60 M. The CST from the BECT showed a cesium loading of $7.18x10^{-3}$ $mmol_{Cs}/g_{CST}$ or 21.7 Ci_{Cs-137}/kg_{CST}. It also showed loading of iron, and calcium, though less than Batch 1A (See Tables II and III for competitor loading).

TABLE I. Summary of Sample Results for Qualification

		Batch 1	**Batch 1A**	**Batch 2**
Supernate Characteristics	Density [g/mL]	1.089	1.164	1.161
	Na [M]	2.02	3.79	3.60
	NO_2^- [M]	N/A	0.0701	0.0538
	Total Cs* [mg/L]	N/A	1.51	1.57
Cesium Loading on CST	Cs** [$mmol/g_{CST}$]	N/A	$8.96x10^{-3}$	$7.18x10^{-3}$
	Cs-137*** [Ci/kg_{CST}]	N/A	23.2	21.7

*Total cesium calculated from inductively coupled plasma- mass spectrometry results
**Cesium measured on the CST from the BECT
***Loading predicted by ZAM model at concentrations of Cs shown by the supernate sample

Overall Processing Parameters

Batch 1A

Batch 1A processed 575,000 L (152,000 gal) of radioactive salt solution from Tank 10H between January 16, 2019 and February 15, 2019. Two IXCs were used in a lead-lag configuration with IXC 1 in the lead position and IXC 2 in the lag position. The flow rate through TCCR was set to 30 L/min (8 gpm) for startup and lowered to 19 L/min (5 gpm) once all the air had been pushed out of the system. The optimal flow rate for IXC performance based on qualification results is 19 L/min (5 gpm). No TCCR system induced shutdowns occurred during Batch 1A, although other operational concerns or events resulted in TCCR shutdown. Seven of these operational shutdowns occurred throughout Batch 1A processing. Examples include lowering of the transfer pump in Tank 10H and retrieving samples from Tank 11H. Final shutdown occurred due to loss of transfer pump suction.

Batch 2

Batch 2 processed 220,000 L (58,000 gal) of radioactive salt solution between June 21 and June 29, 2019. Two IXCs were used in a lead-lag configuration with IXC 2 in the lead position and IXC 3 in the lag position. The flow rate through the TCCR process was set to 30 L/min (8 gpm) for startup and lowered to 19 L/min (5gpm) once all the air had been pushed out of the system. The flow rate remained there for the majority of the batch. It was raised to 30 L/min (8 gpm) and 38 L/min (10 gpm) for a couple hours each on June 25 to observe system performance. It was raised to 30 L/min (8gpm) again on June 27 for a longer duration of approximately 7 hours. These increases showed corresponding increases in IXC differential pressure and decreases in duration between prefilter backflushes. No operational shutdowns occurred during Batch 2 processing. Final shutdown occurred due to transfer pump shutdown from an indeterminate cause. Restart was not recommended due to the low volume of supernate feed remaining in the tank and the short duration between filter swaps.

Prefiltration Performance

The TCCR unit contains two dead-end prefilters arranged in parallel. The filters consist 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 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. 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 across the filter, time between filter swaps, and recovery of flow and differential pressure following a backwash.

Batch 1A

The backflush setpoint was a differential pressure of 24 kPa (3.5 psi) for at least 2 min. The backflush was performed at a flow rate of approximately 28 L/min (7.5 gpm) for 30 s. Typical durations between filter flushes were 2 hr at the beginning of processing. This decreased to approximately 0.5 hr by the end of the batch. This decrease could be attributed to the concentration of solids in Tank 10H as the majority of the waste was treated and sent to Tank 11H, but is difficult to quantify. It could also be due to the short duration of the backflush which does not clear the plenum of all solids. When flow is returned to the filter, these remaining smaller sized solids are deposited directly on the filter media. Differential pressure recovery after backwashing was around 4.1 -5.5 kPa (0.6-0.8 psi) at the beginning of Batch 1A. This saw minor degradation to 4.8 – 6.9 kPa (0.7-1.0 psi) by the end of the batch. A typical trend of prefilter performance over Batch 1A is shown in Figure 2.

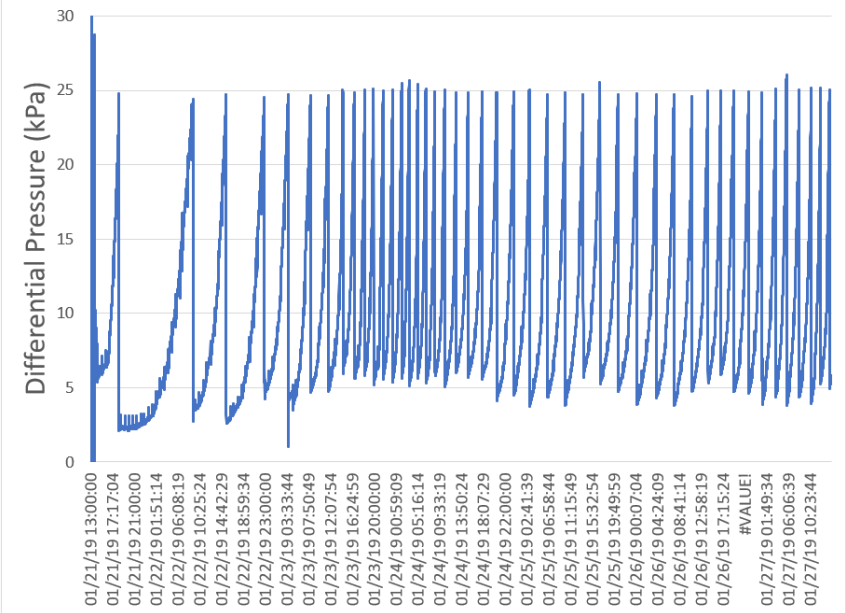

Fig. 2. Prefilter Performance over a Week of Processing Batch 1A.

Batch 2

The backflush setpoint was a differential pressure of 34 kPa (5 psi) for at least 2 min for Batch 2. This was increased due to the smaller batch size and higher concentration of sodium and solids compared to Batch 1A. The backflush was performed at a flow rate of approximately 1.5x the flow rate through the system. The duration was 30 s for approximately the first half of the batch. This was increased to 1 min for the remainder of the batch. Time between filter swaps was approximately 1 hr at the beginning of Batch 2 processing and decreased to around 0.08-0.12 hr by the end of the batch. Differential pressure recovery following backwashing saw some more minor degradation from 4.8-6.9 kPa (0.7-1.0 psi) to 11.7-13.1 kPa (1.7-1.9 psi).

The flow rate was increased from 19 L/min (5 gpm) to 30 L/min (8 gpm) and 38 L/min (10 gpm) during Batch 2. When the flow rate was raised, the time between filter swaps decreased accordingly, as shown in Figures 3a and 3b.

Figure 3. Prefilter Performance during Processing of Batch 2.

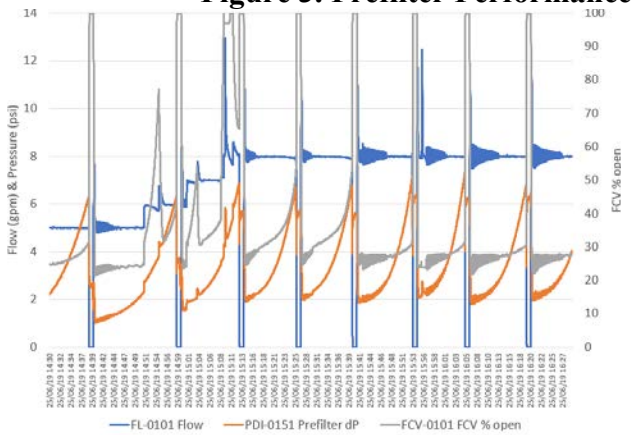

Fig. 3a. Prefilter performance during increase from 5 gpm to 8 gpm.

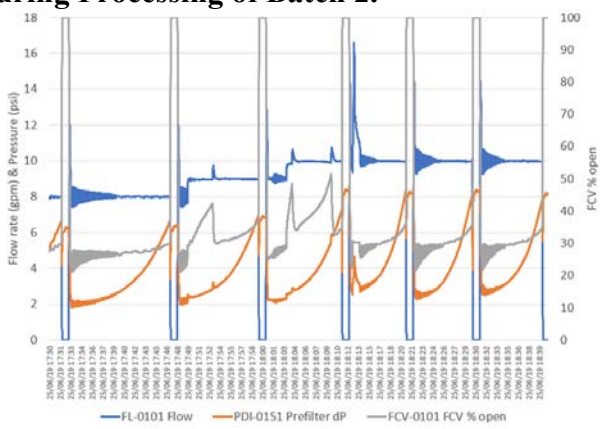

Fig. 3b. Prefilter performance during increase from 8 gpm to 10 gpm

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. Pressure across the IXCs equalize during a prefilter backflush sequence and shows a negative value due to the difference in height between the gauges.

Batch 1A

Columns 1 and 2 were used to process Batch 1A. Figure 4 indicates the hydraulic performance of these IXCs was as expected at a differential pressure of approximately 62 kPa (9 psi). The larger spikes in dP are seen during startup as higher flow rates are used to remove air from the system.

The stable dP readings of 62 kPa (9 psi) indicate there was no loss of media fines or channeling during Batch 1A.

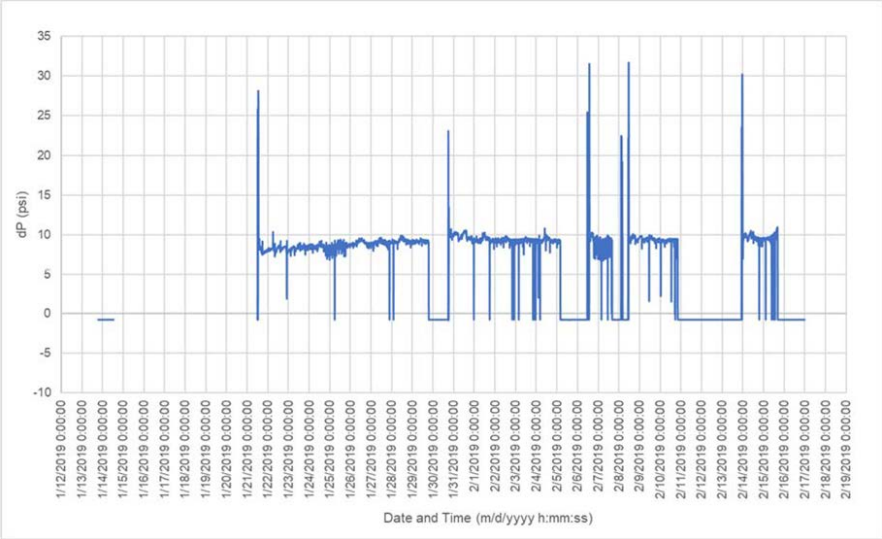

Fig. 4. Differential Pressure Readings Across IXCs for Batch 1A.

Batch 2

Columns 2 and 3 were used to process Batch 2. Figure 5 shows these IXCs did not behave similarly to columns 1 and 2 during processing of Batch 1A under the same flow rates. The configuration used in Batch 2 of Column 2 in the lead position and Column 3 in the lag position showed a much lower dP of approximately 48 kPa (7 psi) at 19 L/min (5 gpm). The feed flow rate was increased during operation of Batch 2. This resulted in corresponding increases in dP across the IXCs. At flow rates of 30 and 38 L/min (8 and 10 gpm), the dPs observed were 159 and 248 kPa (23 and 36 psi).

The dP observed during operation at 19 and 38 L/min (5 and 10 gpm) are lower than expected values, while the dP observed during operation at 30 L/min (8 gpm) is higher than expected from the estimated values for 19 and 38 L/min (5 and 10 gpm). Higher than expected pressures indicate a restriction in flow path, while lower pressures indicate the removal of resistance.

The cause of the discrepancy between Batches 1A and 2 is unknown at this point. These differences will be investigated during processing of Batch 3 to attempt to isolate the cause.

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 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, but these rinses 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. This leaching behavior has not been quantified so that the impact of the sludge heel on the DSS can be discerned.

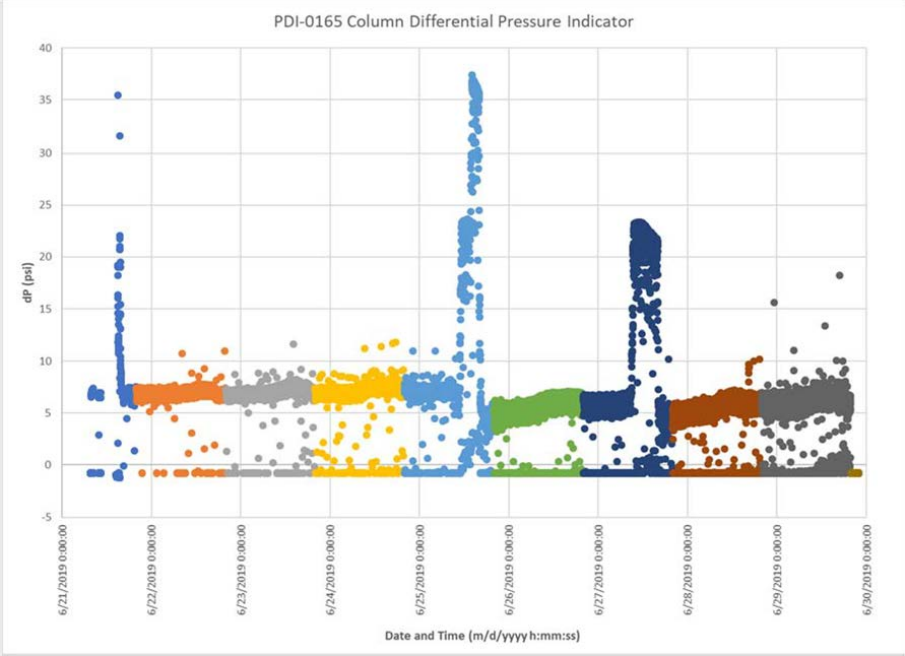

Figure 5. Differential Pressure Readings Across IXCs 2 & 3 for Batch 2.

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.

At this time, there is no accurate means to quantify the effect of leaching soluble radionuclides from the residual sludge heel in Tank 11H in the post-production sample results nor is there an accurate means of determining the true Cs-137 concentration in the DSS from radiation monitors with the current placement of the monitor and recorded data from the operating system. Efforts are underway to improve the ability to understand the performance of the unit by improving detection capability. These efforts include the design of new radiation monitors that have a more sensitive detection limit and can account for varying process flow rates and the varying column

exit location, depending on which IXCs are online. The varying flow rates and column exit locations 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. 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 DF of 1,000. Additionally, both Batches 1A and 2 have met the WAC to be sent to SPF for permanent disposition.

Batch 1A
Sample results for Cs-137, plutonium, and other select ions are shown in Table 2 below. These sample results indicate a DF of about 50, much less than the target DF of 1000. A total of seven in-service IXC flushes were sent to Tank 11H during operation of Batch 1A. These 570 L (150 gal) flushes are required upon restart by the TCCR Operation. The DF is about 70 when these flushes are accounted for at feed concentrations of Cs-137.

The sample results in Table II indicate increases in some species that should have been removed by TCCR or be unchanged. There was a large increase of strontium in the Tank 11H post-production sample. This may be due to leaching of strontium from the sludge heel and/or residual strontium on the tank wall and cooling coils. This increase was also seen in plutonium radiation rates that could only have come from a sludge heel in Tank 11H. Potassium, another known competitor for CST sites, was removed from the feed. The calcium and iron concentrations are higher than expected after the results of the BECT for Batch 1A.

Figures 6 and 7 below show the radiation monitor data across Batch 1A processing. The instantaneous DF for Batch 2 measured by the radiation monitors was greater than 5,000 for all flow rates. Dips in the inlet feed radiation rates are indicative of process shut-downs (Figure 6). Radiation levels in the resin trap (post-IXC filtration) and DSS line spike concurrently with IXC flushes (Figures 7 & 8). Based on the low radiation readings near the resin trap, no solids or CST accumulated in the resin trap.

TABLE II. Tank 11H Post-Production Results vs. Batch 1A Feed

Analyte	Tank 11H Pre-Production	Tank 11 Post-Production	Tank 10H Batch 1A Feed	% Difference of Feed and Tank 11H Post
Specific Gravity	1.001	1.164	1.164	0
Cs-137 [dpm/mL]	$1.76x10^{6}$	$1.13x10^{6}$	$4.887x10^{7}$	-97.7
Sr-90 [dpm/mL]	$4.34x10^{5}$	$2.07x10^{6}$	$3.16x10^{5}$	555
Pu-238 [dpm/mL]	$1.69x10^{3}$	$5.79x10^{3}$	$3.32x10^{4}$	-82.6
Pu-239/240 [dpm/mL]	$8.08x10^{1}$	$3.20x10^{2}$	$3.78x10^{2}$	-15.3
Pu-241 [dpm/mL]	$3.82x10^{2}$	$1.09x10^{3}$	$2.79x10^{3}$	-60.9
K^{+} [M]	$<1.15x10^{-4}$	1.70x10-3	$2.21x10^{-3}$	-23.1
Ca^{2+} [M]	$6.15x10^{-5}$	$<3.52x10^{-5}$	$7.14x10^{-5}$	-50.7
Fe^{3+} [M]	$7.77x10^{-6}$	$<5.53x10^{-5}$	$4.99x10^{-5}$	10.8

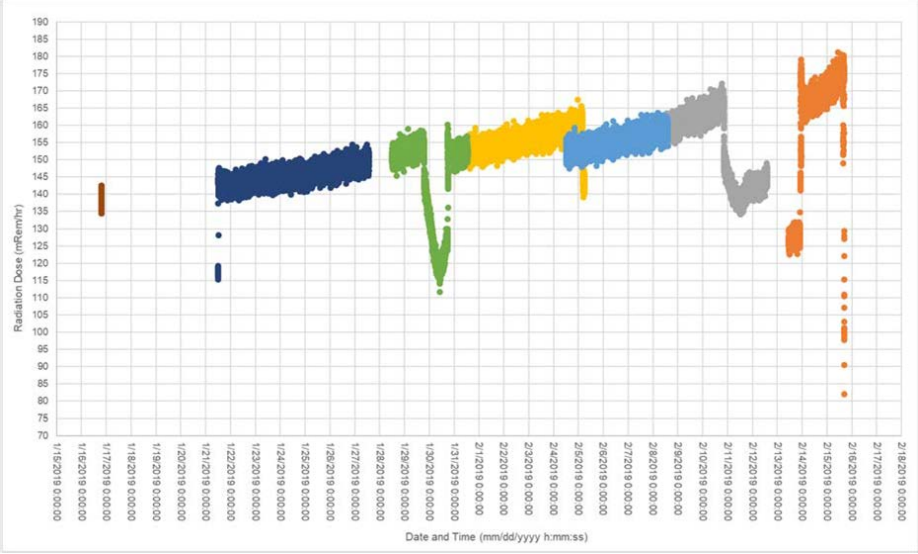

Figure 6. Radiation Monitoring for Inlet Feed during Batch 1A.

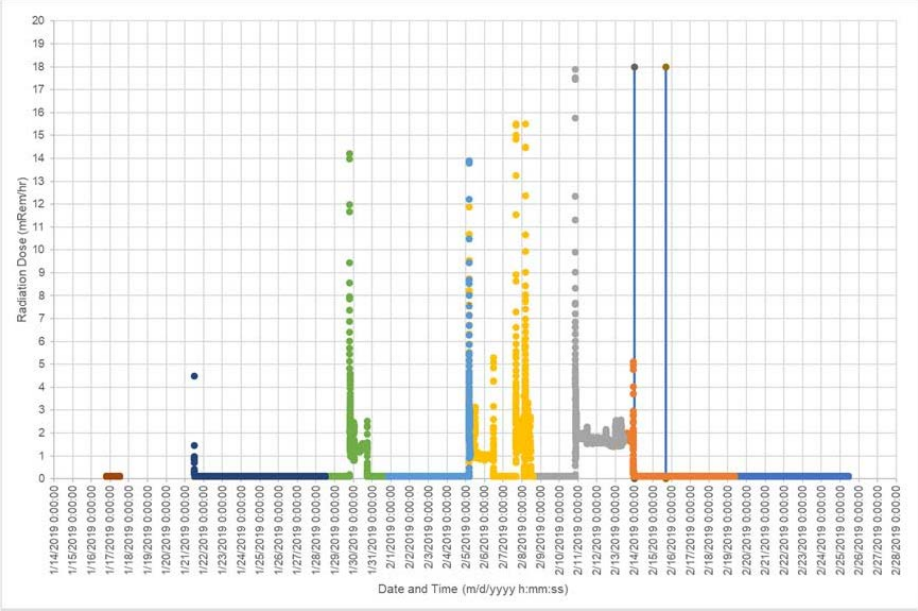

Figure 7. Radiation Monitoring for the DSS to Tank 11H.

Batch 2

Prior to Batch 2, a couple of evolutions were completed in Tank 11H: a water rinse was performed to reduce the soluble radionuclides in the sludge heel and a caustic addition was made for corrosion control. This caustic addition caused a large increase in Cs-137 concentrations. This large increase of cesium can be attributed to leaching of the Tank 11H sludge heel into the supernate.

Sample results for Cs-137, plutonium, and other select ions are shown in Table III. These sample results indicate a DF of about 170, less than the target DF of 1000. One IXC flush was sent to Tank 11H to begin operation of Batch 1A. This 570 L (150 gal) flush was required upon

restart with a previously used column in the lead position by the TCCR Operation. The DF is about 300 when these flushes are accounted for at feed concentrations of Cs-137.

The minor differences in the composition, as seen in Table III, could be attributed to impurities introduced by the caustic addition, leaching from the sludge heel, and/or the caustic addition suppressing a majority of the anions apart from hydroxide. The introduction of DSS to Tank 11H caused a chemistry change that may have resulted in some of the radioisotopes leaching from the sludge heel, resulting in a higher concentration than the feed. The increase of Sr-90, Pu-230/240, and Pu-241 in the Tank 11H post-production sample is largely attributed to this phenomenon. This may help explain why the DF is biased low compared to the DF of the radiation monitors.

Figures 8 and 9 below show the radiation monitor data across Batch 2 processing. The instantaneous DF can be determined by comparing the inlet and outlet DF. The instantaneous DF for Batch 2 measured by the radiation monitors was greater than 4,000 for all flow rates. Dips in the inlet feed radiation rates are indicative of process shut-downs (Figure 8). Increases in levels are indicative of increased feed flow rates.

TABLE III. Tank 11H Post-Production Results vs. Batch 1A Feed

Analyte	Tank 11H Pre-Caustic Addition Surface Sample	Tank 11H Pre-Caustic Addition Depth Sample	Tank 11H Pre-Production	Tank 11H Post-Production	Tank 10H Batch 2 Feed	% Difference 100*(Tk 11 Post -Tk 10)/Tk 10
Specific Gravity	1.013	1.033	1.149	1.152	1.159	-0.6
Cs-137 (dpm/mL)	1.62×10^{5}	8.31×10^{5}	1.88×10^{6}	8.28×10^{5}	5.10×10^{7}	-98
Sr-90 (dpm/mL)	NM	NM	NM	3.09×10^{6}	6.39×10^{5}	380
Pu-238 (dpm/mL)	NM	NM	NM	7.73×10^{3}	3.08×10^{4}	-74
Pu-239/240 (dpm/mL)	NM	NM	NM	4.17×10^{2}	4.05×10^{2}	1.9
Pu-241 (dpm/mL)	NM	NM	NM	2.00×10^{3}	1.93×10^{3}	3.8
K^{+} (M)	$<1.40\times10^{-3}$	$<1.40\times10^{-3}$	NM	4.4×10^{-3}	9.65×10^{-4}	354
Ca^{2+} (M)	7.11×10^{-5}	$<4.39\times10^{-5}$	NM	4.74×10^{-5}	2.84×10^{-5}	67
Fe^{3+} (M)	$<2.19\times10^{-5}$	$<2.18\times10^{-5}$	NM	2.38×10^{-5}	$<5.01\times10^{-6}$	>370

*Items marked NM were Not Measured during that particular sampling analysis.

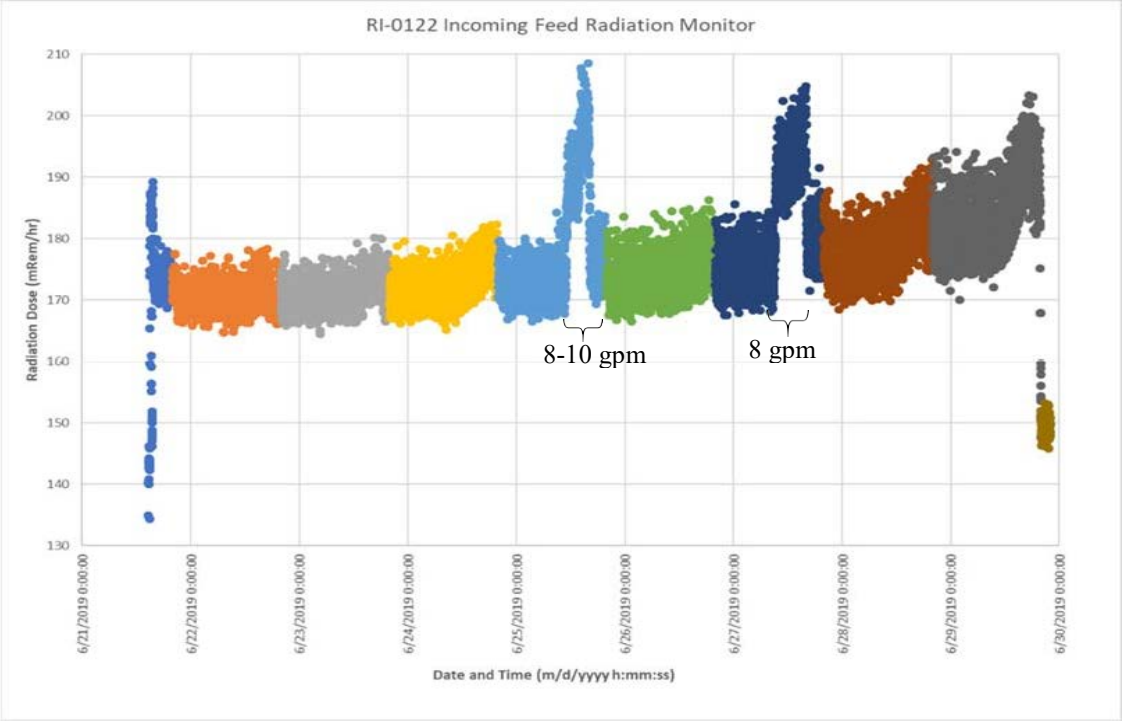

Figure 8. Radiation Monitoring for Inlet Feed during Batch 2.

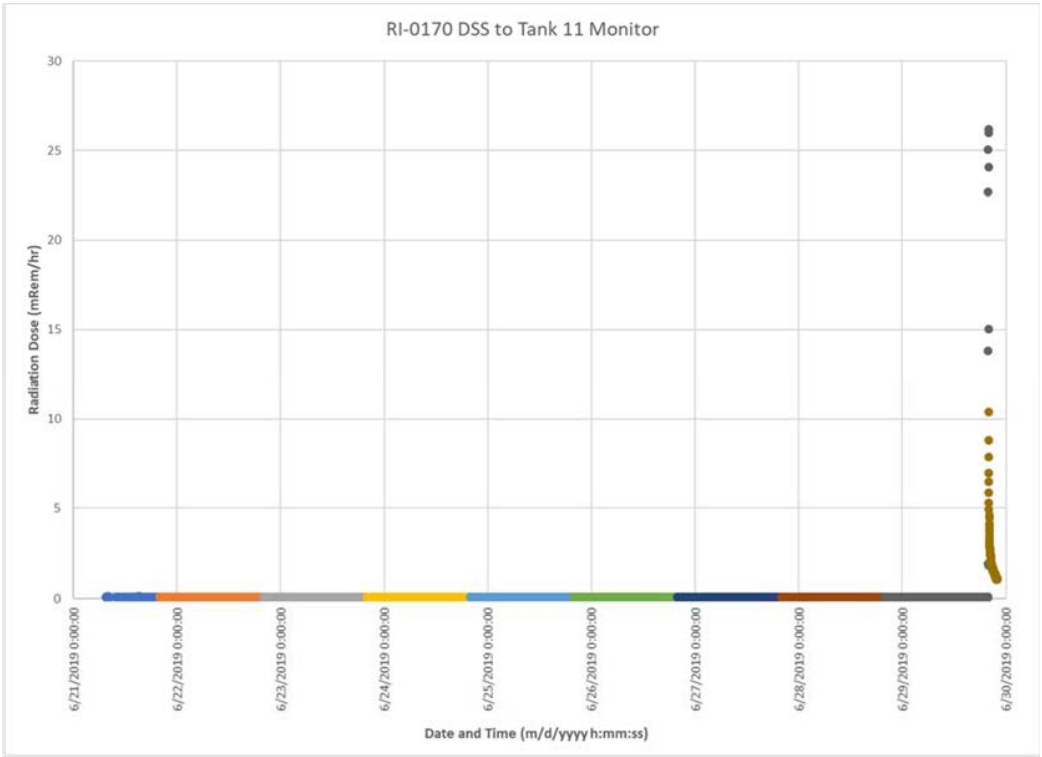

Figure 9. Radiation Monitoring for the DSS to Tank 11H during Batch 2.

OPTIMIZATION FOR PROCESSING TANK 9H WASTE

Work is underway at SRS to expand TCCR operation to include radioactive salt waste in Tank 9H. The waste will be dissolved in Tank 9H and transferred to Tank 10H where it will be qualified for processing through the TCCR unit. This process is referred to as TCCR 1A. Some physical changes to systems can be made between the TCCR demonstration on Tank 10H waste and TCCR 1A.

The CST chosen for the TCCR demonstration is the hydrogen form of CST, R9120B, manufactured by UOP-Honeywell. This form of CST has been engineered into a bead using a binder material. Different commercially available forms of the CST beads are being investigated in an effort to increase Cs loading on the CST. Concurrently with investigating a possible different bead size of CST, new columns are being procured for TCCR 1A with a 2.54 cm (1in) reduced diameter (diameter of ~48 cm or 19 in) from those used during the TCCR demonstration (diameter of ~51 cm or 20 in). The dimensions of the shielding cask around each column will remain the same. This increases the annulus space between the shielding and the column itself. This annulus space has been filled with water to promote the transfer of decay heat out of the IXC. Improved heat transfer will allow for increased loading of cesium on the IXCs without boiling within the IXC. This increased cesium capacity of the CST will have to be balanced with protecting the values used in the safety basis accident analysis calculations.

The prefilters performed well during the TCCR demonstration but could be improved. New prefilter assemblies are being procured to fit within the installed prefilter housings with an increased surface area of 0.68 m^2 (7.33ft^2), instead of 0.54 m^2 (5.81 ft^2). This is being done by increasing the number of filter tubes in the bundle, while decreasing the diameter of each tube. This increase and redesign of the tube bundle will reduce the available plenum space where solids could potentially reside. Additionally, the absolute filtration rating of the filters may be decreased from 10 µm and will be constructed using a different filter media. This change in mesh size should allow the solids within SRS waste to form a filter cake on top of the filter media that will itself act as a filter and remove the smaller particles that may be plugging the current filter media. The different media, support structure, and guard design will improve the cleaning of the filters during backflushes. This will also be aided by increasing the filter backflush duration to two minutes to ensure all of the smaller particle size solids are completely flushed from the filter housing.

Determination of instantaneous DF of the TCCR system is another large improvement being pursued for TCCR 1A. This may include radiation monitors external to the system placed underneath the transfer lines at the inlet and outlet of the TCCR unit. These monitors would be more sensitive than the currently installed monitors and have the ability to account for the fact that Ba-137m, which is measured by the monitor, is not yet in secular equilibrium with Cs-137. This factor will be based on distance of the radiation monitor from the exit of the last, online IXC and flowrate through the TCCR system. The overall DF, determined by sampling the DSS in Tank 11H, will be investigated during processing of TCCR Batch 3. Further investigation will be performed, if required.

CONCLUSIONS

TCCR has successfully processed 795,000 L (210,000 gal) of radioactive salt solution from Tank 10H. There has not yet been a system induced shutdown, though seven operational shutdowns occurred during processing of Batch 1A to collect data and lower the transfer pump. The prefilters performed as expected at all flow rates with only minor degradation in recovery of differential pressure after a backflush sequence. The time between backflushes decreased as each batch reached the end of processing as solids concentrated in Tank 10H. The hydraulics in the IXC mostly performed as expected at all flow rates. There is one column that did not perform as expected and will be investigated further during Batch 3 processing. The overall bulk average DF is estimated to achieve the minimum design requirement bulk average DF of 1,000. The resulting DSS met all of the SPF WAC requirements and will be sent to SPF for permanent disposition.

The radiation levels at the resin trap do not indicate any solids or CST accumulated. The minor levels after Batch 2 can be attributed to residual salt solution and/or background radiation within the enclosure.

Future batches of Tank 10H material will investigate the leaching rate of constituents into the DSS in Tank 11H by increasing the sampling frequency and improve the accuracy of the readings from the current radiation monitors or the addition of new radiation monitors. They will demonstrate flow rates below 19 L/min (5 gpm), 1 and 3 IXC performance, and an increased backflush duration to minimize prefilter degradation.

REFERENCES

1. T.B. Caldwell, “Tank Closure Cesium Removal (TCCR) System”, X-SOW-H-00002, Rev. 4, March 2017.
2. L.A. Britanisky, T.L. Fellinger, & M.K. Keefer, “Understanding and Controlling Cesium Loading on Crystalline Silicotitanate – 19601,” WM2019 Conference Proceedings, March 3-7, 2019.
3. N.J. Eigenbrot, “Safety Basis Development for the Tank Closure Cesium Removal (TCCR) Demonstration – 19596,” WM2019 Conference Proceedings, March 3-7, 2019.
4. M.T. Keefer, W.P. Mayson, III, G.C. Arthur, & T.L. Fellinger, “Tank Closure Cesium Removal at Savannah River Site- 19604,” WM2019 Conference Proceedings, March 3-7, 2019.