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Cesium-137 Measurements of Spent CST Columns from the Savannah River Site Tank Closure Cesium Removal (TCCR) Project (22360)

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ABSTRACT

Savannah River National Laboratory (SRNL) personnel were asked to assay large lead-shielded columns filled with crystalline silicotitanates (CST)in support of the Savannah River Site Tank Closure Cesium Removal (TCCR) Project. With an overall length of 366 cm and a diameter of over 86 cm, the columns contain a CST "bed' 270 cm long and 50 cm in diameter. They are shielded with 12.7 cm of lead and a total of 3.5 cm of stainless steel. The CST columns are used to capture Cs-137 as waste is transferred from one tank to another (1,2,3,4,5,6).

Since the vast majority of the activity on the column was expected to be Cs-137, there were a wide variety of possible detector choices. For reasons of equipment availability and ease of analysis, the measurement was performed with an ISOCS(*In Situ* Object Counting System)^{TM}-characterized HPGe detector. For many measurements, it was difficult to get close to the column. With a 30-degree collimator, the field of view was so large, only three vertical slices were taken. In addition, four columns were stored on a single concrete pad, with only moderate spacing between them. Without the thick lead shielding and the proximity of other columns, this would have been a straightforward measurement. There were approximately 6 tenth-value layers of lead in the columns, and this made the analysis quite complicated. Trying to measure the columns expected to contain lower levels of activity without interference from the "hotter" columns was also a challenge. The difficulties and results of the measurements will be discussed.

INTRODUCTION

Personnel from the Nuclear Measurements Group at Savannah River National Laboratory were tasked with assaying four large heavily shielded columns filled with CST. These columns were stored on a concrete pad near one of the operational areas at the Savannah River Site (SRS). Approximately 3.66 m long and 86 cm in diameter, the columns contained a 2.7 m CST "bed" 50 cm in diameter. The activity contained in the column was shielded with 12.7 cm of lead and a total of 3.5 cm of stainless steel.

These columns were used as part of an accelerated cleanup demonstration project called Tank Closure Cesium Removal (TCCR). In short, tank waste is pumped through the columns on the way from one tank to another. The CST is designed to collect cesium. This will remove nearly all the Cs-137 from the tank waste. This decontaminated salt solution will eventually be processed at the Saltstone Production Facility at SRS and turned into Saltstone Disposal Units.

Over 1.1 million liters (300,000 gallons) of tank waste were processed through the four columns under study. As the Cs-137 levels are at least 100 times that of any other radionuclide, no radiation signature should be expected, apart from the 662 kV gamma ray from Cs-137/Ba-137m.

Column Design

Each CST bed is 270 cm long and 50 cm in diameter, with a 0.95 cm thick stainless-steel shell. The effective bed volume is 495 liters, and it is loaded with 515 kg of CST. This leads to a bulk density of 1.04 kg/l of CST.

Each CST bed is placed within a shield constructed of stainless steel and lead. With 1.27 cm stainless cylinders inside and out, the main shielding consists of a 12.7-cm thickness of lead poured between the two stainless-steel layers. The bulk thickness of the shielding was roughly confirmed with Co-60 transmission dose-rate measurements.

The top and bottom of both the shielding and the column are somewhat more complicated than a simple cylinder. For the purposes of measurements taken looking at the center, cylindrical portion of the columns, the complexity of the ends will be ignored in modeling. Figure 1 shows isometric views of the CST bed and the outer shielding.

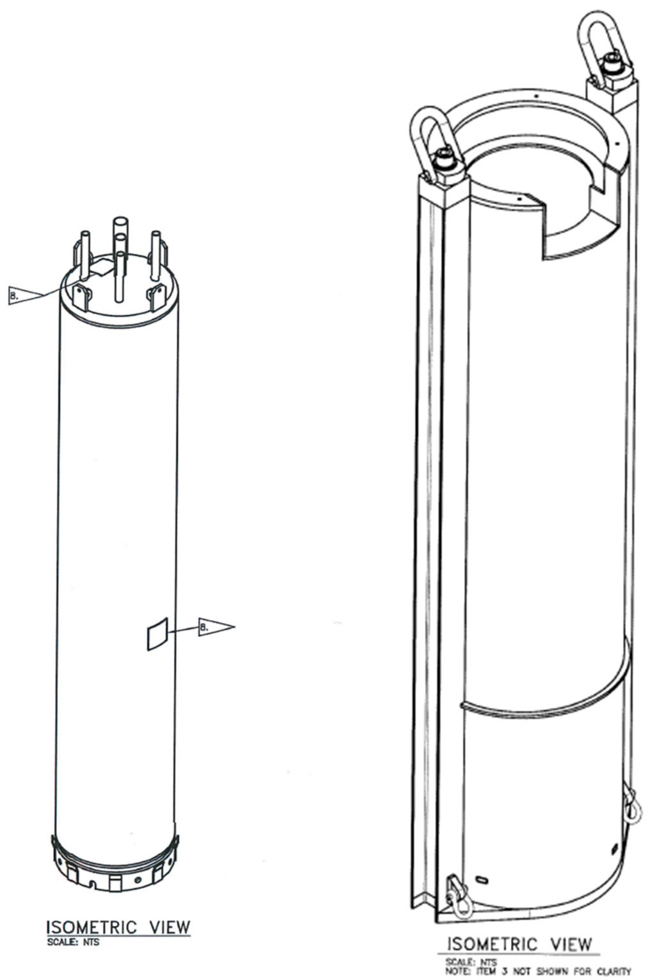

Figure 1: Isometric views, not to scale, of the CST column (left) and the outer shielding (right)

Though the columns may absorb Sr-90 [7] and even some actinides, in addition to the Cs-137, the only radiation signature expected to be seen is the 662 keV gamma-ray from Cs-137/Ba-137m. This makes it possible to use quite a few different measurement techniques. A low-resolution technique would have sufficed, but because of equipment availability and ease of analysis, it was decided to use an ISOCS^{TM}-characterized HPGe detector with a 30-degree lead collimator.

In early 2021, the four columns of interest were moved from the operational area to a remote storage location and set, in pairs, on a concrete pad. The columns within a pair are separated by about 2.5 meters and pairs are separated by about 4 meters. The entire pad is fenced for radiation concerns. Figure 2 below shows a photograph of the pad.

Figure 2: Photograph of four TCCR columns on a fenced concrete pad

METHODS

ISOCS (*In Situ* Object Counting System) [8] is a combination of hardware, software, and services from Mirion Technologies that allows the user to calculate, rather than measure, the absolute counting efficiency of a wide variety of geometries. For a simple geometry, or for one that can be effectively simplified, this allows for a much smaller analysis burden. The user does not need to make large simplifying assumptions to use point/line/plane approximations, nor is it necessary to build complex geometry models using a code such as Monte Carlo N-Particle (MCNP). ISOCS integrates directly into the GENIE 2000 [9] suite of gamma-analysis software from Mirion, providing additional efficiency.

The CST column geometry, especially that portion to be measured, is easily represented as an arrangement of concentric cylinders and, in that way, is easy to model and calculate with the ISOCS software. The 12.7 cm lead shield makes highly sensitive gamma-ray absorption calculations key but that is likely to be a problem with any modeling method.

The column measurements were performed with an ISOCS-characterized HPGe detector from Mirion Technologies. While it is possible to use the ISOCS methodology with a generic detector profile, using a detector for which the response has been characterized by Mirion generally leads to smaller uncertainties. The HPGe detector used was coaxial with a 7.6 cm diameter endcap and a relative efficiency of at least 30%. To shield the detector, a 2.5 cm thick collimator with a 30-degree field of view was used. The collimator is available as an option within the ISOCS calibration software, and this is another time-saving measure.

Access to the columns was restricted by the fencing that can be seen in Figure 2. For some columns, the preferred geometry required a detector distance of approximately 3 meters. With a 30-degree field of view, it was impossible to assay small "slices" of the column from that distance. It was decided to use 3 vertical measurements heights at each distance. Ideally, these heights would have been evenly spaced over the column height (the CST column height). Operational difficulties precluded this and unfortunately the top two measurements were closely spaced.

Measurements were taken for 600 seconds at each of the three heights in a series, in addition to a background performed at the middle height, two vertical series were done for each column. From dose rate measurements of the columns, it was known in advance that the column labeled #1 was by far the hottest and that columns #3 and #4 were contained far less activity. Because of this, and the wide field of view of the collimator at long distances, efforts were made to prevent the measurement of a "cold" column with a "hot" column in the view of the detector. Figure 3 shows a simple diagram of the column and detector positions for the measurements.

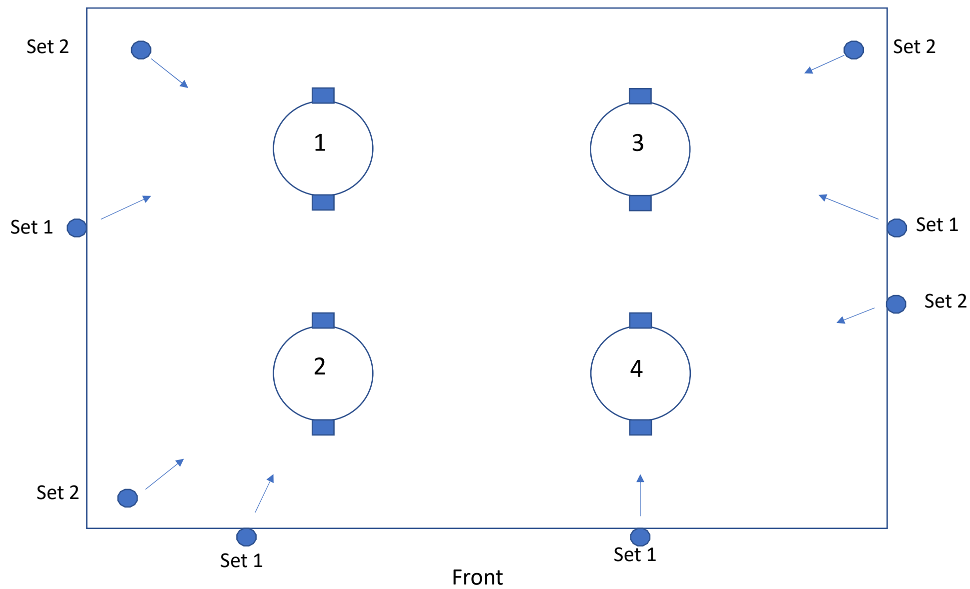

Figure 3: Layout of the columns and the measurement positions (not to scale)

Because of the layout of the land surrounding the pad, it was not possible to measure from the back side of the pad, as shown in the drawing. Figure 2 shows the encroaching forest on the far side of the pad. This made it difficult, perhaps impossible, to measure column #3 without column #1 or column #2 in the shot. How this is accommodated in the analysis will be described in the discussion below.

Once the measurements were taken, it was necessary to build efficiency models using the Mirion Geometry Composer software that ships as part of the ISOCS package. Using measurements from "as-built" drawings, a general geometry was constructed that, with adjustments to the detector distance, could be used for each measurement series. Figure 4 below shows a wireframe drawing of the detector and column. This drawing represents the highest measurement point at the farthest distance.

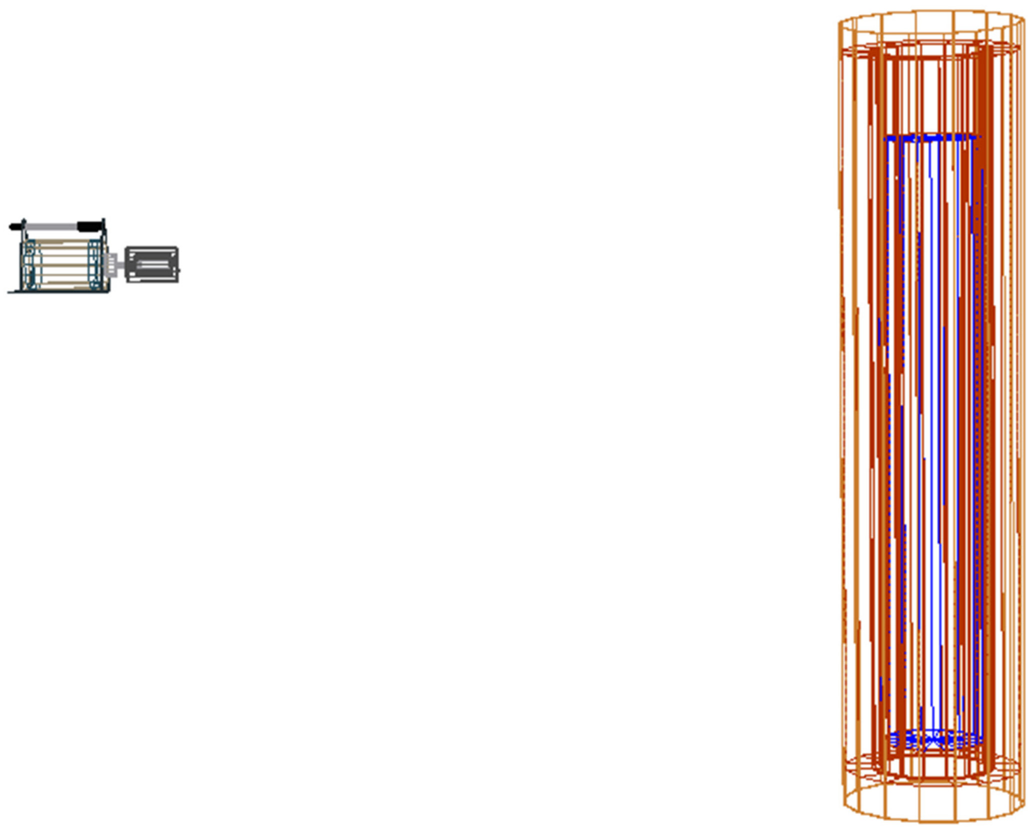

Figure 4: Wireframe drawing of the detector and shielded CST column

The section of the drawing highlighted in blue represents the CST bed. By modifying the geometry of the source, it is possible to determine the sensitivity of the detector to gamma rays emitted from a specific region of the column. This will be important later in the unfolding discussion. Once the geometry has been constructed within the software, it is straightforward to calculate the efficiency for a particular arrangement.

The goal of these measurements was two-fold, to calculate the total inventory of each column and to determine the vertical distribution of activity on the column. The large field of view of each measurement point makes this somewhat more difficult. A more complicated method must be used.

With three vertical measurements points, it makes sense to calculate the efficiency of the measurement for three separate sections of the column. In this way, a square (3x3) matrix can be assembled representing the efficiency of the three measurement points for each of the three sections of the column. Inverting the matrix allows for the calculation of the activity by section from the three detector count rates. This is shown in Figure 5.

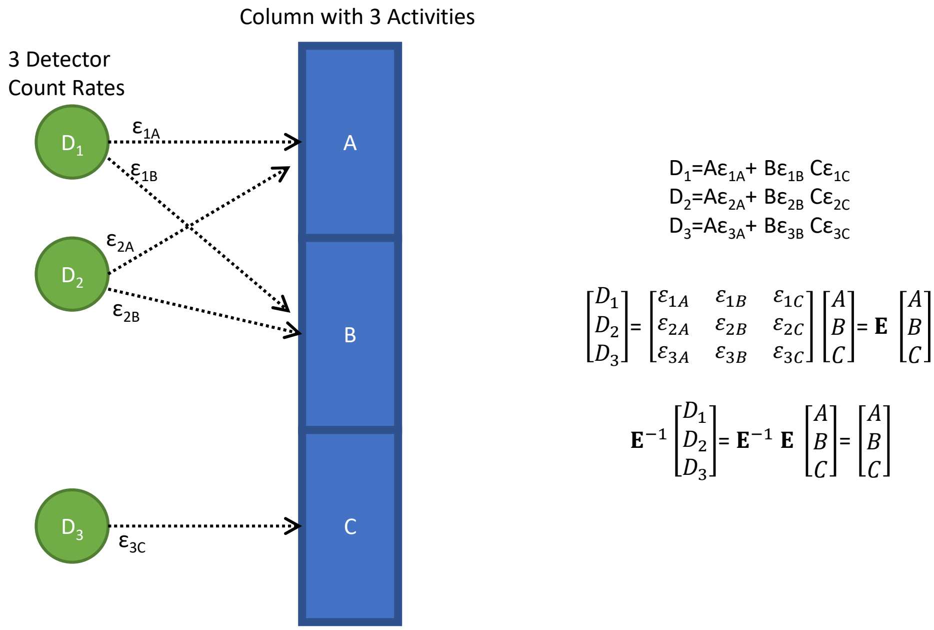

Figure 5: Diagram of the three-section matrix inversion process.

Ideally, the measurement points would have been equally distributed vertically, but this was difficult to achieve in the field and the top and middle measurements were closer than would be ideal. The large field of view of the detector at each location causes a large amount of overlap for all the measurement points. A plot of the geometry for small slices around the center of the CST column (0 on the X axis) is shown in Figure 6. This demonstrates the overlap well. This overlap surely degrades the quality of the matrix inversion method.

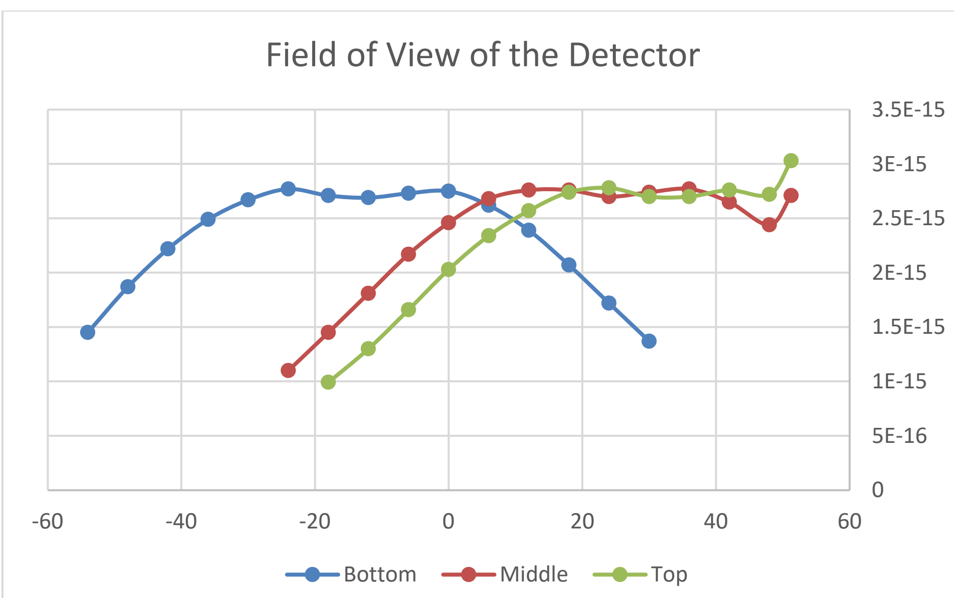

Figure 6: The efficiency of the detector positions for Column #1 in small slices around the center (0 on the X axis).

DISCUSSION

In the simplest analysis, one could imagine taking the middle measurement point and using it to calculate the total inventory of the column. It does not view the entire column, but the analysis can adjust for that fact. If this is done for the very first measurement set on Column #1, one gets a result over four times higher than the total maximum activity for all four columns. If the analysis is done using the matrix inversion method, the total for the Column #1 is nearly three times the theoretical maximum. Results for the other three columns are just as unlikely as these, but Column #1 was the hottest, by far, and should have yielded the most accurate, uncontaminated results.

At this point, the analysis was suspended, and a full review was initiated. The detector and associated electronics are maintained within a regular Quality Assurance (QA) program. It was quickly determined that the detector system was operating within normal parameters.

The next part of the process where significant errors could likely be introduced is within the geometry calculations. This geometry represents a large amount of shielding and perhaps it is beyond the normal capabilities of the code or perhaps a small error in the absorption calculation is compounded repeatedly by the thickness the lead in the problem. It proved a bit more difficult to get a handle on this part of the problem than the detector QA.

The first step that was taken in confirming the geometry was to look at how the ISOCS software handled different thicknesses of shielding. A simple problem was constructed in both ISOCS and MCNP [10] that transmitted gamma rays through lead shielding of various thicknesses to a generic HPGe detector. This was also done manually, using standard absorption calculations. These results were compared and found to be nearly identical, at least for ISOCS and MCNP, for transmission through 12.7 cm of lead. The manual results, which are likely over-simplified, were identical within a reasonable uncertainty.

The next step was to confirm the effect of the actual physical geometry on the efficiency. Small "mockups" of portion of the geometry were built in ISOCS and MCNP. They compared very well. This is what ISOCS was built for, so perhaps this result is unsurprising, if comforting.

Finally, a full model of the geometry was constructed in MCNP to compare to the ISOCS model. To make the problem runnable in a reasonable amount of time, it was not done using full analog capture. The flux per energy at the front surface of the detector, within the collimator, was used as a substitute for the actual signal in the detector. For the purposes of this comparison, the exact capture behavior of the detector was not relevant, and the effect would be similar with any thickness of lead. In the end, these results compared very well also. They were not identical, but that is to be expected and the differences were at least an order of magnitude smaller than the 300 to 400% that would be required to match the scan results to the theoretical maximum.

Have exhausted obvious paths of inquiry regarding the analysis, it was necessary to step back and question the most basic assumptions. Specifically, is the shielding really 12.7 cm of full-density lead? In In consulting with the design engineers, it eventually became clear that while what tolerances were relatively tight for manufacturing this type and size of item, they may allow for the size of the effect that are seen in the analysis. For a nominal 127 mm lead shield, it would require only that it be between 9 and 10 mm undersized to account for the observed problems. It is also true that an equivalently large reduction in the density would have a similar effect. More likely, it is a combination of the two problems.

After the shields were constructed, some non-destructive examination was done to evaluate the shielding provided. A 1.25 TBq Co-60 source was used to measure the dose rate through the shield at more than 350 points throughout column, with 16 radial measurements on 23 vertical planes. Over the whole column, the standard deviation of these measurements was approximately 25%, but within any one vertical plane, the results may vary by a factor of three.

It was mentioned earlier that both measurement series for Column #3 were contaminated by other "hot" columns in the field of view. This made is very difficult to put a value on the activity for this column. The best approximation that could be achieved was to calculate the results for column #3 Set 1 and subtract the results of Column #4 set 2 (which should have similar contamination levels). This led to a more reasonable result for Column #3, but only at the cost of much larger errors.

RESULTS

All totals are relative to the predicated value for each column. The predicted values are arrived at by calculating the potential inventory of the four columns and then distributing that activity among the columns based on the operating conditions and the measured dose rates.

For the Matrix Inversion Method (MIM), results for the top, middle and bottom show the percentage of the total and the total is relative to the predicted value for a column. For the Full Column Approximation (FCA), results are shown relative to the predicted value for each measurement and the total is the mean of the three values, relative to the prediction.

Col.			MIM	FCA		Col.			MIM	FCA
		Top	40%	515%				Top	40%	1550%
#1		Middle	60%	505%		#2		Middle	60%	1590%
Set 1		Bottom	0%	333%		Set 1		Bottom	0%	890%
		Total	310%	450%				Total	720%	1350%
			MIM	FCA					MIM	FCA
		Top	0%	560%				Top	10%	610%
#3		Middle	25%	1200%		#4		Middle	90%	800%
Set 1		Bottom	75%	6100%		Set 1		Bottom	0%	325%
		Total	4600%	2600%				Total	0%	580%

CONCLUSIONS

This initial effort to assay these CST columns for absolute activity values has proven far more difficult than originally anticipated. The difficulty lies not within the bounds of the nuclear measurement process. Determining the true thickness and density of a large, manufactured lead shield is the trouble. One could imagine beginning nuclear measurements much earlier in the process and using a large source to measure the shield thickness before it is even loaded with the CST column. This would allow for the calculation of an effective thickness/density for a measurement.

Failing this, it is hard to imagine how one could measure the total inventory of these columns with any reasonable uncertainty. The thickness of the lead likely varies enough to affect the measurement significantly. With careful planning and execution, it should be possible to achieve a good relative measurement of the column activities (assuming similar thicknesses for the shield, which may not be correct) and scale this to the maximum expected inventory.

REFERENCES

[1] Chew, D. P.; Hamm, B. A.; Wells, M. N. Liquid Waste System Plan Revision 21. January 21,2019 SRR-LWP-2009-0001, Rev. 21 https://www.srs.gov/general/pubs/srr-lw-systemplan.pdf

[2] King, W. D., Hamm, L. L., Hang, T. L., and Fondeur, F. F. *CST Batch Contact Equilibrium Testing of TCCR Tank 10H Batch 1 and 1A Waste Supernate Samples*. United States: N. p., 2019. Web. doi:10.2172/1510920

[3] K. M. L. Taylor-Pashow, *Summary of Analytical Results from Tank 11H Tank Closure Cesium Removal (TCCR) Post-Batch 2 Processing Sample*, SRNL-L3100-2019-00028, Rev. 0, August 15, 2019.

[4] K. M. L. Taylor-Pashow and C. A. Nash, *Summary of Expedited Results from Samples Supporting Tank Closure Cesium Removal (TCCR) Batch 3*, SRNL-STI-2020-00269, Rev. 0, July 2020.

[5] K. M. L. Taylor-Pashow, T. Hang, C. A. Nash, and T. B. Edwards, *Summary of Expedited Results from Samples Supporting Tank Closure Cesium Removal (TCCR) Batch 1 and Modeling Results for Cs Loading on CST*, SRNL-L3100-2018-00102, Rev. 3, March 6, 2019

[6] Beck, M., Duncan R., Gammon, T. *Technology Basis for Savannah River Tank Closure Cesium Removal Project – 17448.* WM2017 Conference, March 5-9, 2017, Phoenix, Arizona, USA

[7] Marsh, S. F.; Svitra, Z. V.; Bowen, S. M. Distributions of 14 Elements on 60 Selected Absorbers from Two Simulant Solutions (Acid-Dissolved Sludge and Alkaline Supemate) for Hanford HLW Tank 102-SY; Los Alamos Report, LA-12654 {UC9401, October 1993.

[8] Geometry Composer [Version 4.4.1] (November 1, 2016), Mirion Technologies Inc, Meriden CT.

[9] Genie2000 [Version 3.4.1] (November 1, 2016), Mirion Technologies Inc, Meriden CT.

[10] C. J. Werner(editor), "MCNP Users Manual – Code Version 6.2", Los Alamos National Laboratory, report LA-UR-17-29981 (2017)