

Cesium Exchange onto Crystalline Silicotitanate from Blended Hanford Tank Wastes

October 2021

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Summary

The Tank Side Cesium Removal (TSCR) system was developed to filter and remove cesium (Cs and ¹³⁷Cs) from Hanford tank waste supernate in preparation for vitrification. The Cs removal will be conducted with crystalline silicotitanate (CST) ion exchange media. Under the planned waste-processing strategy, the tank waste supernate will be queued for TSCR processing in tank 241-AP-107 (AP-107). Once AP-107 tank waste volume is sufficiently depleted, the waste supernate from tank 241-AP-105 (AP-105, the holding tank before transfer to AP-107) will be transferred to tank AP-107. Supernate from another tank will be transferred to the holding tank, AP-105, for eventual transfer to tank AP-107. These supernate streams will undergo blending in tanks AP-107 and AP-105; the volume blend ratios will be driven by how much the tank waste supernate volumes are depleted before the next tank waste is added. The consequence of tank waste blending on Cs uptake by CST was of interest and was tested via batch contacts; results are reported herein.

Testing was conducted on AP-107, AP-105, and 241-AW-102 (AW-102)¹ tank wastes that were first processed through CST ion exchange beds to strip ¹³⁷Cs (and Cs), reducing the radiation dose and allowing the matrices to be contact-handled by laboratory personnel during this testing. Tank waste volume blend ratios represented the upper and lower bounding volume blend ratios, 21.8 and 3.8, respectively, along with an intermediate blend ratio, 12.8. (Note that a larger value of the volume blend ratio represents a larger difference between the two volumes blended, so the blend ratio of 21.8 results when the maximum amount of supernate has been removed from the feed tank, leaving minimum heel, before supernate from the holding tank is added.) Cs isotherms were then developed for each tank waste blend at two different temperatures (15.7 and 24.3 °C). The blended tank waste results were compared to the unblended tank wastes. Small (10% to 15%) increases in Cs loading Q values (mmoles Cs per g CST) were observed at 15.7 °C for the blended tank wastes, but no substantive changes in Q values were observed at 24.3 °C. Q values agreed within 6% relative standard deviation for a given set of three waste blends at a given temperature. Table S.1 summarizes the Q values for each tank waste blend and temperature. None of the tank waste blends exceeded the TSCR documented safety analysis (DSA) limit for ¹³⁷Cs loading (141,600 Ci, corresponding to 238 Ci ¹³⁷Cs/kg CST).²

Unblended AW-102 tank waste was processed to determine Q values at three process temperatures (13.4, 15.7, and 24.3 °C). The Q value vs. temperature curve fit was similar to those previously reported,³ where $Q = -0.00226 \times T + 0.1008$ (corresponding to a DSA threshold at 1.4 °C process temperature, T). Therefore, AW-102 tank waste processing is not expected to exceed the DSA ¹³⁷Cs loading limit even if temperature drops to 13.4 °C.

¹ AW-102 tank waste was identified as a suitable surrogate of other tank wastes that could be processed via TSCR.

² Anderson K. 2020. *Tank Side Cesium Removal Safety Basis Requirement Decision Document*. RPP-RPT-62225, Rev. 0. Washington River Protection Solutions, Richland, Washington.

³ Fiskum SK, EL Campbell, RA Peterson, and TT Trang-Le. 2021. *Temperature Effect of Cesium Exchange onto Crystalline Silicotitanate in AP-107 and AP-105 Hanford Tank Wastes and Two Simulants*. PNNL-31355, Rev. 0; RPT-DFTP-029, Rev. 0. Pacific Northwest National Laboratory, Richland, Washington.

Table S.1 Summary of Q Values with CST Lot 2002009604 and Blended Tank Waste

Process Temp. (°C)	Matrix	Cs (M) ^(a)	Q (mmoles Cs/g CST)	Relative Standard Deviation
15.9	AP-107 tank waste ^(b)	6.91×10^{-5}	0.0868	NA
21.0			0.0790	NA
15.9	AP-105 tank waste ^(b)	5.65×10^{-5}	0.0582	NA
21.0			0.0543	NA
26.0	AW-102 tank waste	4.58×10^{-5}	0.0440 ^(c)	NA
15.7	96% AP-105, 4% AP-107	5.65×10^{-5}	0.0702	2.1%
	93% AP-105, 7% AP-107	5.68×10^{-5}	0.0692	
	79% AP-105, 21% AP-107	5.80×10^{-5}	0.0721	
24.3	96% AP-105, 4% AP-107	5.65×10^{-5}	0.0472	3.3%
	93% AP-105, 7% AP-107	5.68×10^{-5}	0.0469	
	79% AP-105, 21% AP-107	5.80×10^{-5}	0.0498	
15.7	96% AW-102, 4% AP-105	4.63×10^{-5}	0.0677	2.9%
	93% AW-102, 7% AP-105	4.65×10^{-5}	0.0690	
	79% AW-102, 21% AP-105	4.80×10^{-5}	0.0652	
24.3	96% AW-102, 4% AP-105	4.63×10^{-5}	0.0443	5.4%
	93% AW-102, 7% AP-105	4.65×10^{-5}	0.0484	
	79% AW-102, 21% AP-105	4.80×10^{-5}	0.0490	
15.7	96% AP-105, 4% AW-102	5.57×10^{-5}	0.0689	1.4%
	93% AP-105, 7% AW-102	5.54×10^{-5}	0.0688	
	79% AP-105, 21% AW-102	5.40×10^{-5}	0.0706	
24.3	96% AP-105, 4% AW-102	5.57×10^{-5}	0.0502	2.0%
	93% AP-105, 7% AW-102	5.54×10^{-5}	0.0483	
	79% AP-105, 21% AW-102	5.40×10^{-5}	0.0498	
13.4	AW-102 tank waste	4.58×10^{-5}	0.0706	NA
15.7			0.0649	NA
24.3			0.0459	NA

(a) June 21, 2021, reference date.

(b) Fiskum SK, EL Campbell, RA Peterson, and TT Trang-Le. 2021. *Temperature Effect of Cesium Exchange onto Crystalline Silicotitanate in AP-107 and AP-105 Hanford Tank Wastes and Two Simulants*. PNNL-31355, Rev. 0; RPT-DFTP-029, Rev. 0. Pacific Northwest National Laboratory, Richland, Washington.

(c) The Q value was calculated from data reported in Rovira AM, SK Fiskum, JR Allred, JGH Geeting, HA Colburn, AM Carney, TT Trang-Le, and RA Peterson. 2019. *Dead-End Filtration and Crystalline Silicotitanate Cesium Ion Exchange with Hanford Tank Waste AW-102*. PNNL-28783, Rev. 0; RPT-TCT-003, Rev. 0. Pacific Northwest National Laboratory, Richland, Washington.

NA = not applicable

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The authors thank Amy Westesen and Andrew Carney for helping collect temperature profile data, and Renee Russell for conducting the technical review of numerous calculation files, test data packages, and this report. The authors also thank Bill Dey for the quality reviews of the calculation files and this report and Matt Wilburn for his technical editing contribution to this report.

Acronyms and Abbreviations

CST	crystalline silicotitanate
DSA	documented safety analysis
GEA	gamma energy analysis
PNNL	Pacific Northwest National Laboratory
QA	quality assurance
R&D	research and development
RPD	relative percent difference
TSCR	Tank Side Cesium Removal
WRPS	Washington River Protection Solutions, LLC
WWFTP	WRPS Waste Form Testing Program

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1.0 Introduction

Ion exchange columns containing crystalline silicotitanate (CST)^{1,2} will be used to remove cesium (and ¹³⁷Cs) from Hanford tank waste supernates to start early vitrification operations at the Waste Treatment and Immobilization Plant. Per the Washington River Protection Solutions, LLC (WRPS) planned waste-processing strategy, tank waste supernate will be queued for Tank Side Cesium Removal (TSCR) processing in tank 241-AP-107 (AP-107). Once the AP-107 tank waste volume is sufficiently depleted from processing through TSCR, the waste supernate from the tank 241-AP-105 (AP-105, the holding tank before transfer to AP-107) will be transferred to tank AP-107. Supernate from another tank will be transferred to the holding tank, AP-105, for eventual transfer to tank AP-107. These supernate streams will undergo blending and the extent of blending will be driven by how much the tank waste supernate volumes are depleted during the course of processing.

The supernate volume left in tank AP-107 after processing supernate through TSCR is estimated to range from 52 to 246 kgal.³ The total fluid capacity in tank AP-107 is estimated to be 1184 kgal (total tank capacity of 1247 kgal minus a 63-kgal sludge heel). Therefore, a range of blending volumes is envisioned for normal processing operations. The projected maximum and minimum tank waste supernate volume blending ratios that could be introduced to TSCR are shown in Table 1.1 (prototypically represented by AP-105 mixed into AP-107). The minimum/maximum supernate heels represent the minimum/maximum volumes of supernate remaining in the TSCR feed tank (AP-107) before the next tank waste supernate (from holding tank AP-105) is transferred to the feed tank. Thus, minimum and maximum volume blend ratios were calculated by dividing the added waste volume by the supernate heel volume.

Table 1.1. Projected Tank Waste Blending and Volume Ratios

Supernate Heel Description	Heel AP-107 (kgal)	Added AP-105 (kgal)	AP-105/AP-107 Volume Ratio
Minimum	52	1132	21.8
Maximum	246	938	3.8

The TSCR documented safety analysis (DSA) established a maximum ¹³⁷Cs column loading limit of 141,600 Ci (238 Ci ¹³⁷Cs/kg CST) (Anderson 2020). The maximum of 0.10 mmoles Cs/g CST or distribution coefficient (K_d) of 1470 mL/g at the nominal feed condition of 6.8×10^{-5} M Cs, Cs isotopic distribution of 20 wt% ¹³⁷Cs, and a column volume of 596.6 L CST was established as the maximum load conditions (Fiskum et al. 2021a). Achieved Cs loading is driven by a confluence of factors, including but not limited to:

- CST production lot (Cs capacity has been shown to vary with different production lots) (Fiskum et al. 2019a)
- TSCR processing temperature (the lower the temperature, the higher the Cs loading; Zheng et al. 1996; Fiskum et al. 2021b)

¹ CST is produced by Honeywell UOP, LLC (Des Plaines, IL) under the product name IONSIV™ R9140-B.

² CST is a Nb-substituted silicotitanate formulated by staff at Texas A&M University and Sandia National Laboratories and then manufactured in an engineered spherical form (Braun et al. 1996). Its chemical and physical properties, column dynamics, temperature tolerance, and radiation tolerance were previously described in a literature review (Pease et al. 2019).

³ Personal communication from Benjamin Gallaher (WRPS) to Reid Peterson (PNNL), March 22, 2021.

- Contact time with CST (longer contact time enhances Cs exchange) (Fiskum et al. 2019b, 2020)
- ¹³⁷Cs isotopic mass fraction (the lower the mass fraction of ¹³⁷Cs, the more total Cs can be accommodated)
- Total Cs concentration in the feed (as Cs concentration increases, mass loading increases)
- Matrix effects (increased carbonate concentration improves Cs exchange; Fondeur et al. 2000) and competitors in the tank waste feed (K may consume Cs exchange sites; Miller et al. 1998)

Extensive testing at Pacific Northwest National Laboratory (PNNL) has been conducted to evaluate the CST efficacy for removing cesium from Hanford tank wastes. Column testing was conducted with waste samples from tanks AP-107 (Westesen et al. 2021a; Fiskum et al. 2019a), AP-105 (Fiskum et al. 2021c), and 241-AW-102 (AW-102; Rovira et al. 2019) at a small scale, mimicking many of the TSCR system operating parameters. However, the CST was not fully loaded with Cs in these small-scale column tests because of feed volume limitations and evolving processing parameters. Total load capacity at the equilibrium feed condition was better assessed through a series of batch contact tests. The AP-107, AP-105, and AW-102 tank waste batch contact tests were conducted at hot cell temperatures (generally >25 °C). Recent testing showed that bounding the worst-case matrix and test conditions geared to maximize Cs (¹³⁷Cs) exchange onto CST will exceed the DSA ¹³⁷Cs loading limit (Fiskum et al. 2021a). More-focused batch contact tests with post-column processed AP-105 and AP-107 tank waste supernates were conducted at several temperatures, including temperatures well below the ambient temperature of the hot cell. The DSA limit was exceeded while processing AP-107 at 12.7 °C; however, processing AP-107 at 15.9 °C and higher did not challenge the DSA limit. Results from batch contact processing of AP-105 tank waste showed the DSA limit would not be challenged even at 12.7 °C. AW-102 batch contact testing was not conducted at low temperature; however, at the hot cell temperature of 26 °C, the K_d value was nominally 1000 mL/g (Rovira et al. 2019); this corresponded to a Q value of 0.0440 mmoles Cs per g CST, which is below the DSA limit.

Testing described herein evaluates the effect that tank waste supernate blending will have on the CST exchange capacity for Cs. These tests used actual Hanford tank waste from which Cs had been removed by ion exchange and thus reduced the radiation dose to research staff by removing radioactive Cs. Stable Cs was added back to the matrices such that isotherms could be developed. Cesium spike concentrations were selected to closely bracket the feed Cs concentration conditions. Thus, these tests incorporated the complex anion compositions expected in actual tank waste matrices and best reflect their effects on Cs exchange limitations onto CST. Blending ratios bounded the upper and lower blending ratios estimated by WRPS staff.

As noted above, compositional variations in TSCR feeds are likely to result in differing total Cs mass loadings to reach the DSA column loading limit of 141,600 ¹³⁷Cs (238 Ci ¹³⁷Cs/kg CST). In this report, the specific blended feed Cs isotopic compositions and total Cs concentrations (assuming no initial Cs removal) were calculated to assess ¹³⁷Cs column loading. Any loading value higher than 238 Ci ¹³⁷Cs/kg CST is defined as an exceedance of the DSA limit for that waste blend.

The specific objectives of this testing are summarized as follows:

1. Create three AP-105 (dominant)/AP-107 mixtures and develop isotherms for the mixtures at 16 and 21 °C.
2. Create three AW-102¹ (dominant)/AP-105 mixtures and develop isotherms for the mixtures at 16 and 21 °C.

¹ AW-102 tank waste supernate is representative of other feeds that could be added to AP-105 supernate.

3. Create three AP-105 (dominant)/AW-102¹ mixtures and develop isotherms for the mixtures at 16 and 21 °C.
4. Compare Cs loading onto CST for different mixtures and unmixed tank waste.
5. Develop Cs isotherms for AW-102 at 13, 16, and 21 °C.

¹ AW-102 tank waste supernate is representative of other feeds that could be added to AP-105 supernate.

2.0 Quality Assurance

All research and development (R&D) work at Pacific Northwest National Laboratory (PNNL) is performed in accordance with PNNL's Laboratory-Level Quality Management Program, which is based on a graded application of NQA-1-2000, *Quality Assurance Requirements for Nuclear Facility Applications* (ASME 2000), to R&D activities. To ensure that all client quality assurance (QA) expectations were addressed, the QA controls of the PNNL's WRPS Waste Form Testing Program (WWFTP) QA program were also implemented for this work. The WWFTP QA program implements the requirements of NQA-1-2008, *Quality Assurance Requirements for Nuclear Facility Applications* (ASME 2008), and NQA-1a-2009, *Addenda to ASME NQA-1-2008* (ASME 2009) and consists of the WWFTP Quality Assurance Plan (QA-WWFTP-001) and associated QA-NSLW-numbered procedures that provide detailed instructions for implementing NQA-1 requirements for R&D work.

The work described in this report was assigned the technology level "Applied Research" and was planned, performed, documented, and reported in accordance with procedure QA-NSLW-1102, *Scientific Investigation for Applied Research*. All staff members contributing to the work received proper technical and QA training prior to performing quality-affecting work.

3.0 Experimental

This section describes the tank wastes used for processing, tank waste blending, CST and CST pretreatment, F-factor determination, batch contact test conditions, and calculations. All work was conducted according to a test plan¹ prepared by PNNL staff and approved by WRPS and a test instruction² prepared by PNNL staff.

3.1 Tank Wastes

Three tank waste supernate samples were collected by WRPS from the Hanford tank farms and delivered to PNNL. At PNNL, the samples were diluted as needed to obtain a targeted 5.6 M Na concentration, filtered, and processed to remove Cs using lead/lag or lead/lag/polish ion exchange columns filled with CST. Table 3.1 provides a crosslinking of tank waste samples and reports describing receipt, dilution, filtration, and Cs removal by ion exchange. The tank wastes processed for blending were obtained from the effluent product following Cs removal in the hot cells. The effluent composite samples were characterized (see Table 3.2), and these compositions were accepted as being representative of the samples selected for blending. The total Cs concentration remaining in each effluent sample was calculated from the ¹³⁷Cs concentration measured by gamma energy analysis (GEA) and the Cs isotopic ratio measured on the original feed sample. The Cs isotopic compositions were corrected after applying decay correction to the ¹³⁷Cs activities to the reference count date (June 21, 2021). In all cases, the residual Cs concentrations in the post-ion-exchanged effluents were approximately four orders of magnitude below the target Cs spike levels (see Section 3.5).

Table 3.1. Processing References for Initial Tank Waste Handling

Tank Waste Supernate	Receipt, Dilution, and Filtration	Cs Removal
AP-105	Allred et al. 2020	Fiskum et al. 2021c
AP-107	Geeting et al. 2019	Westesen et al. 2021b
AW-102	Rovira et al. 2019	Rovira et al. 2019

There was sufficient residual ¹³⁷Cs in the selected AP-107 and AP-105 tank waste effluent samples to effectively trace Cs uptake on the CST. Tracer ¹³⁷Cs (79 µCi) was added to the 600-mL AW-102 tank waste sample aliquot to effectively trace Cs uptake.

¹ Fiskum SK. 2021. Test Plan TP-DFTP-117, Rev. 0.0. *Batch Contact Testing of Blended Tank Wastes with Crystalline Silicotitanate*. Pacific Northwest National Laboratory, Richland Washington. Not publicly available.

² Fiskum SK. 2021. Test Instruction TI-DFTP-119, *Batch Contact Testing of Cs onto Crystalline Silicotitanate in Blends of AP-107, AP-105, and AW-102 Tank Wastes at 16 and 21 °C, and Unblended AW-102 Tank Waste at 13, 16, and 21 °C*. Pacific Northwest National Laboratory, Richland Washington. Implemented June through July 2021. Not publicly available.

Table 3.2. Tank Waste Cesium Ion Exchange Sample Effluent Compositions

Analyte	AP-105 ^(a)	AP-107 ^(b)	AW-102 ^(c)
	TI082-COMP-EFF (M)	TI100-EFF-COMP ^(d) (M)	TCT008-COMP-EFF (M)
Al	5.23E-1	3.74E-1	4.65E-1
As	[9.7E-4]	5.22E-4	<3.5E-4
Ba	<1.3E-6	<8.2E-7	<1.1E-6
Ca	1.02E-3	2.41E-4	[2.3E-4]
Cd	[2.0E-5]	5.96E-5	[1.3E-5]
Cr	6.56E-3	1.09E-2	1.07E-2
Fe	<1.6E-5	2.79E-4	[7.0E-5]
K	1.02E-1	9.54E-2	1.45E-1
Na	6.00E+0	6.20E+0	5.46E+0
Nb	2.89E-5	[2.1E-5]	NA
P	1.44E-2	2.71E-2	2.66E-2
Pb	2.67E-5	<4.8E-5	<3.9E-5
S	4.65E-2	6.94E-2	3.48E-2
Sr	1.64E-7	<8.7E-7	<2.8E-6
Ti	[2.3E-5]	<7.1E-6	[9.3E-6]
U	1.99E-5	[8.4E-5]	5.23E-5
Zn	[4.7E-5]	[3.1E-5]	<3.2E-5
Zr	[4.5E-5]	[2.1E-5]	[2.6E-5]
Total Cs	8.5E-8	6.3E-8	2.6E-10
TCT008-COMP-FEED			
Free hydroxide	1.24E+0	8.9E-1 ^(f)	9.8E-1
F ⁻	<2.6E-4	NA	NA
Cl ⁻	1.10E-1	7.69E-2 ^(g)	NA
NO ₂ ⁻	1.38E+0	1.22E+0 ^(g)	1.16E+0
NO ₃ ⁻	1.89E+0	1.85E+0 ^(g)	1.75E+0
PO ₄ ³⁻	8.72E-3	1.61E-2 ^(g)	2.30E-2
C ₂ O ₄ ²⁻	2.84E-3	5.68E-3 ^(g)	3.57E-3
SO ₄ ²⁻	2.44E-2	1.92E-2 ^(g)	2.51E-2
Total organic C	2.16E-1	1.8E-1 ^(f)	1.92E-1
Total inorganic C ^(e)	4.72E-1	6.5E-1 ^(f)	5.86E-1
¹³⁷ Cs	1.77E-1 μCi/mL	1.45E-1 μCi/mL	5.54E-4 μCi/mL ^(h)
Density	1.285 g/mL	1.271 g/mL	1.26 g/mL

(a) Fiskum et al. 2021c.

(b) Westesen et al. 2021b.

(c) Rovira et al. 2019; free hydroxide, ion chromatography anions, and total organic and inorganic C were measured on the feed sample (TCT008-COMP-FEED) – these component concentrations were not expected to differ in the column effluent.

(d) In the Test Instruction that generated this sample, the sample was labeled “TI100-COMP-EFF”; however, it was inadvertently mislabeled as “TI100-EFF-COMP” in the laboratory analysis documents. The Sample ID shown in the laboratory analysis documents is used in this report to maintain traceability to the analysis results, which are provided in Westesen et al. 2021b.

(e) Assumed to be carbonate.

(f) Fiskum et al. 2019a.

(g) Westesen et al. 2021a.

(h) Before spiking with ¹³⁷Cs tracer; 1.31E-1 μCi/mL after adding tracer ¹³⁷Cs.

NA = not analyzed

3.2 CST Sample and Pretreatment

CST is manufactured by Honeywell UOP, LLC as IONSIV™. WRPS purchased CST product R9140-B,¹ 18 × 50 mesh, lot number 2002009604. PNNL received a sample from WRPS on September 20, 2018, for testing. Delivery and initial subsampling were described previously (Fiskum et al. 2019b).

Batch contact testing was conducted on the full particle size distribution of CST. An 86.0-g aliquot of the as-received CST was collected and rinsed with ~100 mL of 0.1 M NaOH seven times to remove colloidal fines. The CST was then rinsed once with 100 mL of deionized water to remove the bulk of interstitial salt solution. The rinsed CST was set aside to dry at ambient (~20 °C) temperature for 11 days in an open beaker, at which point the CST essentially reached constant mass. Before subsampling, the CST was mixed by hand-tumbling the container.

3.3 F-Factor

Two sets of F-factor samples were collected, each set in duplicate. The first F-factor sample from each set was collected before CST aliquoting into batch contact vials, and the second F-factor sample from each set was collected after CST aliquoting into the batch contact vials. The masses of the F-factor samples were nominally 0.5 g each. The F-factor (dry CST mass per sampled CST mass) was determined after heating CST at two temperatures: one duplicate pair at 105 °C and the other duplicate pair at 427 °C.

For the 105 °C processing, CST samples were placed into tared glass vials, which were then placed into an Isotemp oven (Fisher Scientific Model 280A) set to 105 °C. The vials were periodically removed from the oven and allowed to cool until the glass threads were cool to the touch, but the main vial bodies were still warm. Then the vials were capped and cooled completely, and the gross masses were measured. The net CST masses were calculated. Typically, mass measures were collected every 4 to 12 h until the mass changes were <0.5% between successive drying-weighing events. The F-factors for this duplicate pair (0.9313 and 0.9307) averaged 0.9310 with a relative percent difference (RPD) of 0.07%.

For the 427 °C processing, CST was placed in tared stainless-steel crucibles. The crucibles were transferred into a Thermoline furnace (Fisher Scientific Model FB1415) that was preheated to 427 °C. After 4 h, the crucibles were removed from the furnace and transferred to a desiccator containing moisture-indicating Drierite desiccant for cooling. Then the gross masses were measured, and net CST masses were calculated. Fiskum et al. (2021a) and King et al. (2018) showed that no further mass losses were achieved after heating CST to 427 °C. The F-factors for this duplicate pair (0.8391 and 0.8322) averaged 0.8356 with an RPD of 0.82%.

3.4 Tank Waste Blending

Tank waste blending targeted volume ratios of 21.8 and 3.8 to match the maximum and minimum blend ratios, respectively, that are expected in operations at the tank farms in support of TSCR. An intermediate ratio of 12.8 was targeted as well. The tank waste supernate sample size was measured by mass, and sample volume was determined by dividing the mass by supernate density. A 100-mL final composite volume was targeted to support test objectives. Ideal mixing was assumed. Table 3.3 identifies the blended tank wastes, tank waste volumes, and the achieved volume blend ratios. The blended tank waste identifications are also shown, where A, B, and C indicate the tank waste blend and 1, 2, and 3 indicate

¹ Product R9140-B is provided in the sodium form and did not require conversion from the hydrogen form to the sodium form prior to testing.

the blend ratio. For example, the combination of AW-102 (dominant component) / AP-105 at the intermediate blend ratio of 12.8 is coded C2.

Table 3.3. Tank Waste Blend Preparations and Achieved Volume Blend Ratios

AP-105/AP-107			
Blend ID >>	B1	B2	B3
AP-105	96.03 mL	93.00 mL	78.91 mL
AP-107	4.25 mL	7.06 mL	21.0 mL
Blend density	1.278 g/mL	1.281 g/mL	1.279 g/mL
Volume blend ratio	22.6	13.2	3.76
AW-102/AP-105			
Blend ID >>	C1	C2	C3
AW-102	96.0 mL	93.0 mL	79.0 mL
AP-105	4.61 mL	7.25 mL	20.9 mL
Blend density	1.252 g/mL	1.256 g/mL	1.263 g/mL
volume blend ratio	20.8	12.8	3.77
AP-105/AW-102			
Blend ID >>	D1	D2	D3
AP-105	96.11 mL	93.00 mL	78.99 mL
AW-102	4.39 mL	7.25 mL	20.6 mL
Blend density	1.278 g/mL	1.277 g/mL	1.277 g/mL
Volume blend ratio	21.9	12.8	3.84

Visual inspection after combining the tank wastes revealed no adverse effects associated with blending. Precipitation was not evident, and no gas evolution or separable phases were observed. Duplicate 2-mL aliquots from each blend were collected; their masses were measured to determine solution densities and they were analyzed by GEA to assess the initial ¹³⁷Cs concentrations.

A 300-mL aliquot of AW-102 was held in reserve for batch contact testing in an unblended format.

3.5 Batch Contact Sample Preparation of Blended Tank Wastes

A series of 30-mL polyethylene bottles were spiked with aliquots of 0.0780 M CsNO₃ solution targeting final 1.30×10^{-4} , 3.40×10^{-4} , and 8.75×10^{-4} M Cs concentrations when combined with the tank waste blends.¹ Then 30-mL aliquots of the various tank waste blends were added to the containers with the CsNO₃ spike solution. Added CsNO₃ solution and tank waste blend volumes were determined based on the masses transferred divided by the appropriate solution densities.

Nominal 0.075-g (dry mass basis) aliquots of CST were measured into 20-mL vials (actual masses measured to nearest 0.0001 g). The CST masses were corrected by multiplying by the F-factor (measured at 105 °C). Then a nominal 15-mL Cs-spiked tank waste blend solution aliquot was added; the exact solution volume transferred was determined from net solution mass divided by the blend solution density.

¹ Cesium spike volumes were small relative to the added test matrix volume to minimize matrix alteration from dilution. The spike volumes represented $\leq 1.2\%$ of the added matrix volume.

The remaining ~5-mL headspace in this configuration allowed for significant fluid motion and assured good mixing. The solution volume-to-CST mass phase ratios averaged 200 and ranged from 187 to 219.

Figure 3.1 provides a prototypic process flowchart showing:

- Blending of AP-105 and AP-107 in the 22.6 phase ratio, Blend B1, and comparator sample collection (yellow squares)
- CsNO₃ addition to 30-mL bottles (blue circles)
- AP-105/AP-107 tank waste added to these bottles (yellow/blue ellipses)
- Subdivision of the Cs-spiked AP-105/AP-107 into vials pre-loaded with CST for the contact testing at the specified temperature (green and purple rectangles)

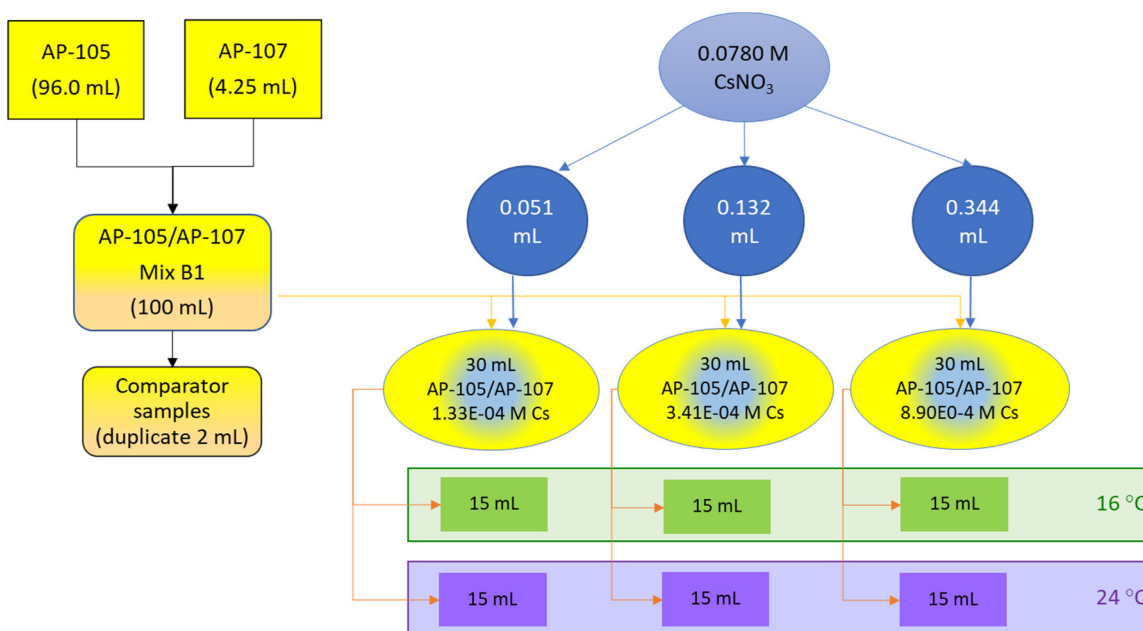


Figure 3.1. Blend B1, AP-105/AP-107 Blending, Cesium Spiking, and Sub-aliquoting for Batch Contacts (prototypic processing)

See Appendix A for specific CST masses, tank waste blend volumes, and starting Cs concentrations.

3.6 Batch Contact Sample Preparation of AW-102 Tank Waste

A series of 125-mL polyethylene bottles were spiked with aliquots of 0.0780 M CsNO₃ solution targeting a final 1.30×10^{-4} , 3.40×10^{-4} , and 8.75×10^{-4} M Cs.¹ Then 90-mL aliquots of AW-102 were added to the containers (with the CsNO₃ spike solution). The added AW-102 tank waste volumes were determined based on the masses transferred divided by the solution density.

Aliquoting of CST and AW-102 into vials was conducted as described in Section 3.5. Each test was conducted in duplicate. The solution volume-to-CST mass phase ratios averaged 199 and ranged from 187 to 219.

¹ Cesium spike volumes were small relative to the added test matrix volume to minimize matrix alteration from dilution. The spike volumes represented $\leq 1.2\%$ of the added matrix volume.

Figure 3.2 provides the process flowchart for AW-102 processing showing:

- ^{137}Cs tracer addition and comparator sample collection
- CsNO_3 addition to 125-mL bottles (blue circles)
- Subdivision of AW-102 into the bottles containing Cs spike solution (yellow/blue ellipses)
- Subdivision of the Cs-spiked AW-102 into vials pre-loaded with CST for the contact testing at the specified temperature (blue, green, and purple rectangles)

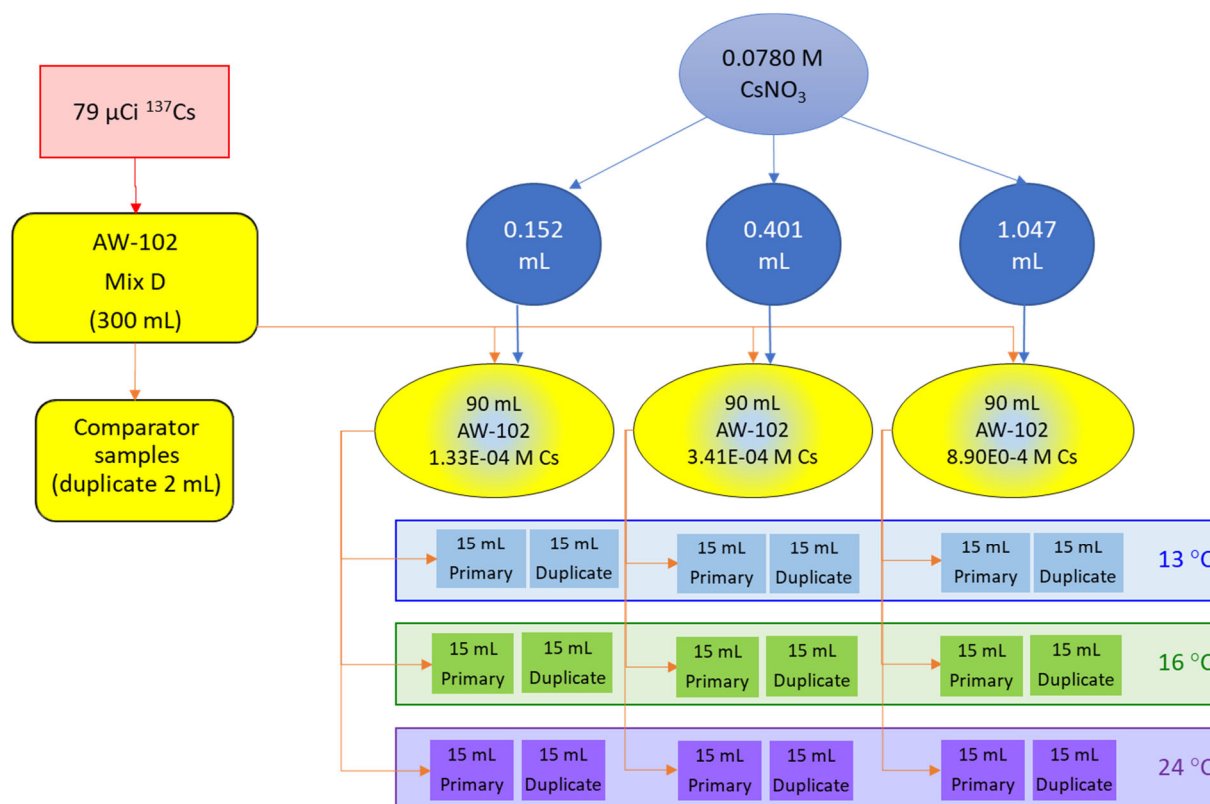


Figure 3.2. AW-102 Subdivision, Cesium Spiking, and Sub-aliquoting (Primary and Duplicate) for Batch Contacts

See Appendix A for specific CST masses, tank waste volumes, and starting Cs concentrations.

3.7 Batch Contact Processing and Analysis

The $24 \text{ }^\circ\text{C}$ test samples were contacted on a MaxQ 2000 (J-Kem Scientific) large orbital shaker with a 19-mm orbit set to 200 rpm. The 13 and $16 \text{ }^\circ\text{C}$ test samples were sequentially contacted in a Benchmark (Sayreville, New Jersey) Incu-Shaker 10LR refrigerated/heated orbital shaker with 19-mm orbit set to 200 rpm.

Vials of water were co-located with each sample set; their temperatures were measured periodically during the contact periods with a type K thermocouple. The water samples served as temperature sentinels for each sample set. Samples were mixed for $\sim 238 \text{ h}$. Figure 3.3 shows the measured test temperatures as a function of contact time. The error bars represent the Type K thermocouple measurement uncertainty of $2.2 \text{ }^\circ\text{C}$. The dashed lines represent the $\pm 2.2 \text{ }^\circ\text{C}$ expected tolerance relative to the target temperature. Table

3.4 provides the weighted mean of the measured temperatures, along with the high and low temperature measures. Clearly, the mixing conducted on the MaxQ 2000 on the benchtop was not highly temperature-controlled. Excess temperature was related to higher room temperature and likely some heat generated from the shaker itself, despite efforts to provide an insulating layer between the sample set and the shaker table. The test plan defined a target temperature of 21 °C. Exceeding the target temperature did not negatively affect assessment of Q vs. temperature but it did challenge direct comparison with tank waste testing previously conducted at 21 °C.

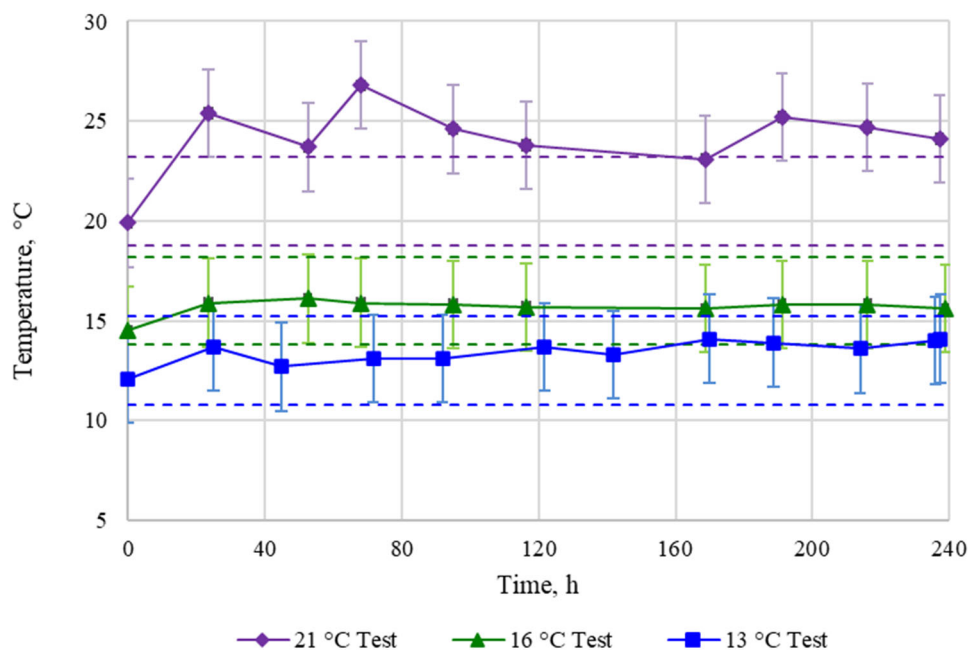


Figure 3.3. Temperature Profiles during Testing for 13 °C, 16 °C, and 21 °C Target Tests

Table 3.4. Contact Temperatures

Target (°C)	Weighted Mean (°C)	Low (°C)	High (°C)
13	13.4	12.1	14.1
16	15.7	14.5	16.1
21	24.3	23.1	26.8

To mitigate the possibility of temperature-driven changes to Cs exchange with increasing (or decreasing) sample temperature during post-contact activities, samples were removed in pairs from the Benchmark Incu-Shaker for final processing. The remaining samples stayed at process temperature and agitation conditions until ready for handling. The samples processed on the MaxQ 2000 shaker were removed as one large group for processing with the assumption that the decrease from 24 °C contact temperature to ~21 °C room temperature would not affect Cs distribution; the maximum stand time at room temperature was <1 h.

The CST settled quickly from the tank waste solution. A nominal 2-mL aliquot was collected and filtered through a 0.45-micron pore size nylon syringe filter. The filtrate was collected in a clean vial for GEA.

The ^{137}Cs was measured in the pre- and post-contacted solutions¹ using GEA, and the measured ^{137}Cs fractionation was used to determine the total Cs exchange. The ^{137}Cs in each sample was analyzed using a high-purity lithium-drifted germanium gamma detector. Comparator samples, prepared from aliquots collected from the starting feed matrix, were measured identically to the post-contacted samples. Sample and comparator count times were adjusted to achieve overall statistical count uncertainties of <1%.

The batch distribution coefficients were calculated according to Eq. (3.1).

$$\frac{(A_0 - A_1)}{A_1} \times \frac{V}{M \times F} = K_d \quad (3.1)$$

where A_0 = initial ^{137}Cs concentration ($\mu\text{Ci/mL}$)

A_1 = final (equilibrium) ^{137}Cs concentration ($\mu\text{Ci/mL}$)

V = volume of the batch contact liquid (mL)

M = measured mass of CST (g)

F = F-factor, mass of the 105 °C dried CST divided by the mass of the undried CST

K_d = batch-distribution coefficient (mL/g)

Final (equilibrium) Cs concentrations (C_{Eq}) were calculated relative to the tracer fraction recovered in the contacted samples and the initial Cs concentration (C_0) according to Eq. (3.2).

$$C_0 \times \left(\frac{A_1}{A_0}\right) = C_{\text{Eq}} \quad (3.2)$$

where C_0 = initial Cs concentration in solution ($\mu\text{g/mL}$ or M)

C_{Eq} = equilibrium Cs concentration in solution ($\mu\text{g/mL}$ or M)

The equilibrium Cs concentrations loaded onto the CST (Q in units of mmoles Cs per gram of dry CST mass) were calculated according to Eq. (3.3).

$$\frac{C_0 \times V \times \left(1 - \frac{A_1}{A_0}\right)}{M \times F \times 1000 \times \text{FW}} = Q \quad (3.3)$$

where Q = equilibrium Cs concentration in the CST (mmole/g CST)

1000 = conversion factor to convert μg to mg

FW = Cs formula weight

The isotherm data were fitted to a Freundlich/Langmuir Hybrid equilibrium fit as previously described by Hamm et al. (2002). The Q values at the specific equilibrium Cs concentrations were calculated from the curve fit.

¹ Both the AW-102 (unblended tank waste) primary and the duplicate samples were processed; the primary samples were measured by GEA; the duplicate samples were analyzed if the primary result appeared to be anomalous. The primary and duplicate samples contacted at 13 °C were measured by GEA.

3.8 Cs Concentrations Expected in Tank Waste Blends

The tank waste feed Cs concentrations expected in neat¹ tank waste materials were calculated to provide reference points for graphing Q vs. Cs concentration. The Cs concentrations and isotopic mass fractions previously reported in tank wastes were used as the initial basis (Fiskum et al. 2019a, 2021c; Rovira et al. 2019). The reported ¹³⁷Cs activity concentrations were decay-corrected to the test count date, June 21, 2021. The revised ¹³⁷Cs activity concentrations were converted to mass concentrations and the sum of ¹³³Cs, ¹³⁵Cs, and ¹³⁷Cs mass concentrations determined. The blended tank waste Cs concentrations were calculated, and the average concentrations are used in graphical representations (Section 4.0). Table 3.5 provides the ¹³⁷Cs mass fractions and total Cs concentrations for the neat tank wastes and the blended tank wastes as of June 21, 2021. The limit of 0.10 mmoles Cs/g CST loading assumed a ¹³⁷Cs mass fraction of 20%; therefore, more Cs may be loaded on the CST given the lower ¹³⁷Cs mass fraction corresponding to the tested matrices.

Table 3.5. Decay-Corrected Cs Concentrations in Tank Wastes, Reference Date June 21, 2021

Tank Waste	¹³⁷ Cs Mass Fraction (%)	Total Cs Concentration ^(a) (M)
AP-105	17.9	5.62E-5
AP-107	19.7	6.51E-5
AW-102	18.5	4.58E-5
AP-105/AP-107 blends		Average 5.71E-5
Blend B1	18.0	5.65E-5
Blend B2	18.1	5.68E-5
Blend B3	18.3	5.80E-5
AW-102/AP-105 blends		Average 4.69E-5
Blend C1	18.5	4.63E-5
Blend C2	18.5	4.65E-5
Blend C3	18.4	4.80 E-5
AP-105/AW-102 blends		Average 5.51E-5
Blend D1	18.0	5.57E-5
Blend D2	18.0	5.54E-5
Blend D3	18.1	5.40E-5

(a) Expected in tank waste before Cs separation.

¹ Before processing through the CST column that removes Cs.

4.0 Results

This section provides the isotherm curves for the blended tank wastes and AW-102 tank waste at each of the process test temperatures. The Cs concentration range for each test was tight, centering on the “linear” portion of the isotherm curve (log-log format) and bracketing the expected Cs feed concentration. Input data supporting the various isotherms and figures is provided in Appendix A.

Figure 4.1 through Figure 4.3 present graphs of the Q values vs. equilibrium Cs molarity for each tank waste blend and test temperature. No substantive change in Q value occurred at a given temperature and within the blend ratios tested. This indicates that the anticipated blending of tank wastes will not result in Cs uptake changes by CST.

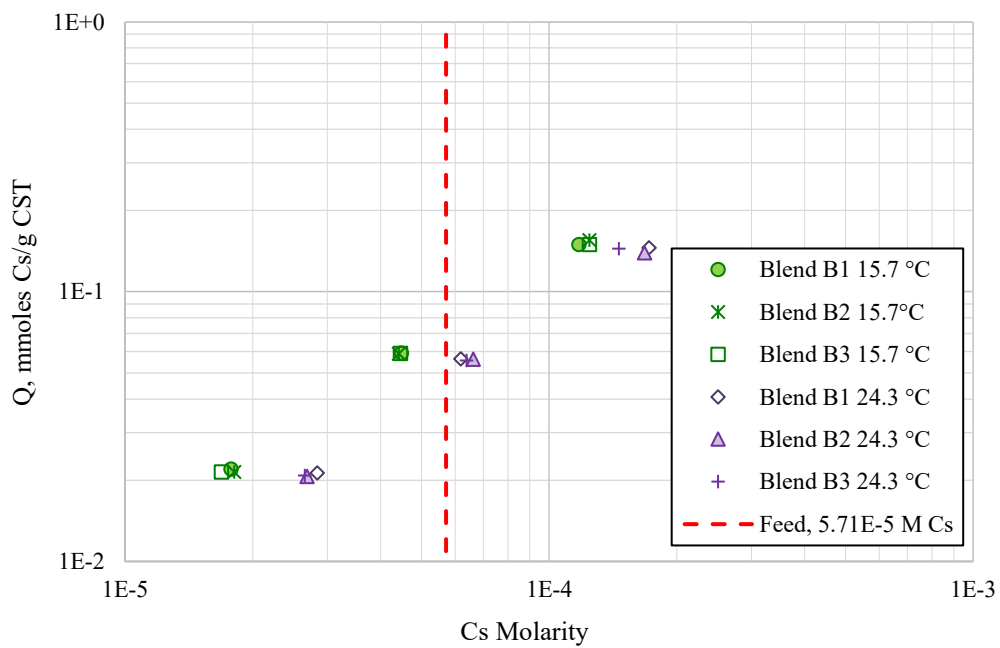


Figure 4.1. Q vs. Equilibrium Cs Concentration, Blends B1, B2, and B3 (AP-105/AP-107 Tank Waste), 15.7 and 24.3 °C

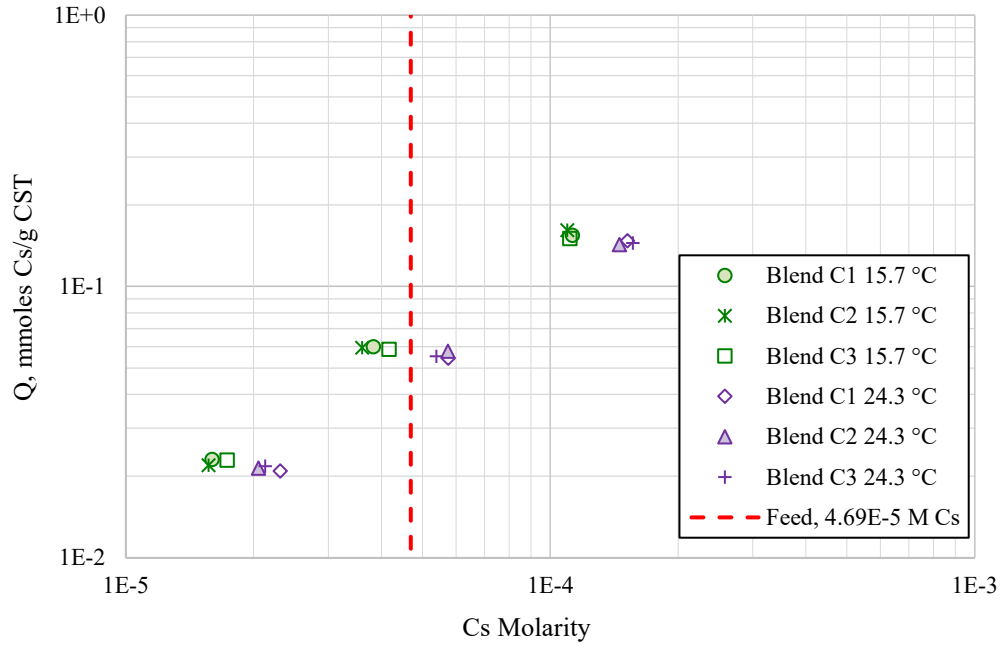


Figure 4.2. Q vs. Equilibrium Cs Concentration, Blends C1, C2, and C3 (AW-102/AP-105 Tank Waste), 15.7 and 24.3 °C

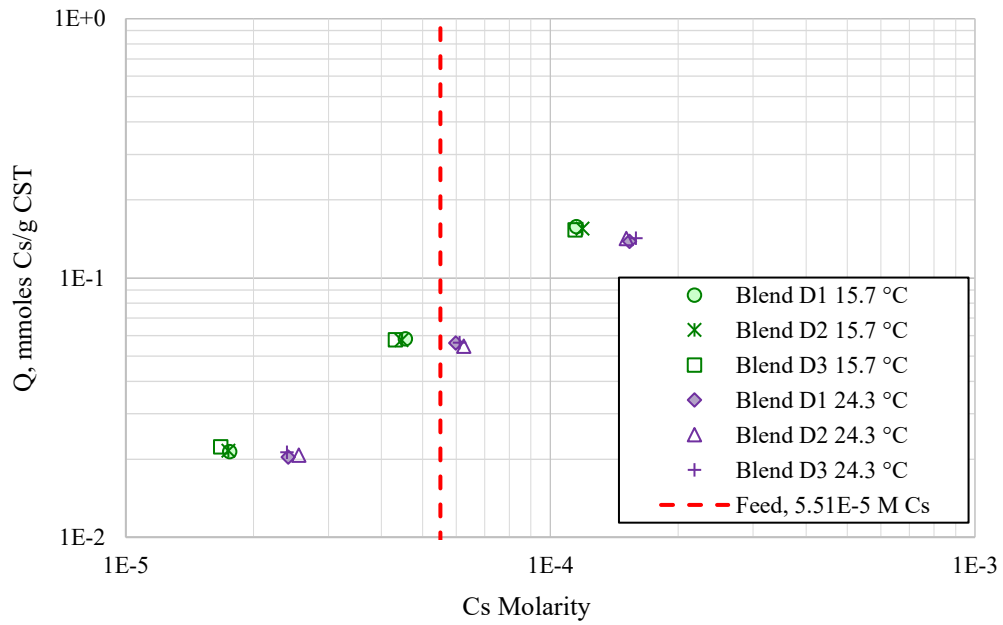


Figure 4.3. Q vs. Equilibrium Cs Concentration, Blends D1, D2, and D3 (AP-105/AW-102 Tank Waste), 15.7 and 24.3 °C

Figure 4.4 presents a graph of Q values vs. equilibrium Cs molarity for AW-102 tank waste at three contact temperatures. The Q value clearly decreases with increasing temperature in agreement with previous studies (Fiskum et al. 2021b). Also shown is the AW-102 (prior to Cs removal in the hot cell) batch contact result determined at 26 °C (Rovira et al. 2019). This result lies on the isotherm established with the Cs-removed AW-102 feed measured at 24.3 °C. The close alignment of the pre-processed AW-102 tank waste result with the post-Cs-removed AW-102 tank waste isotherm demonstrated reasonable comparability. A capacity difference from the ~2 °C discrepancy in test parameters would be expected to drive the pre-processed AW-102 result lower on the graph (decreased Q value); however, this effect is expected to be small.

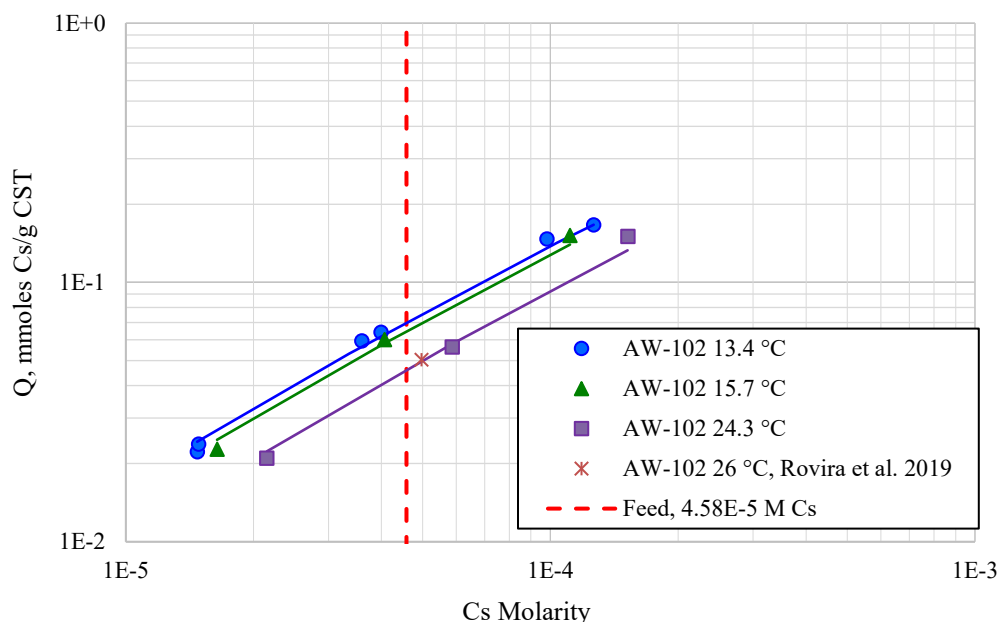


Figure 4.4. Q vs. Equilibrium Cs Concentration, AW-102 at 13.4, 15.7, and 24.3 °C with Freundlich/Langmuir Hybrid Equilibrium Fits, and AW-102 at 26 °C

Hamm et al. (2002) applied the Freundlich/Langmuir Hybrid equilibrium model shown in Eq. (4.1).

$$Q = \frac{\alpha_i \times [Cs]}{(\beta + [Cs])} \quad (4.1)$$

where [Cs] = equilibrium Cs concentration, mmoles/mL or M
 Q = equilibrium Cs loading on the CST, mmole Cs per g CST
 α_i = isotherm parameter constant (mmoles/g), equivalent to total capacity in the matrix
 β = isotherm parameter constant (mmoles/mL or M), selectivity coefficient, dependent on matrix and temperature; the larger the value, the less selective the CST is for Cs (Hamm et al. 2002)

The current testing did not employ a high Cs concentration to determine α_i . However, the maximum capacity was not expected to change significantly from small matrix changes associated with tank waste blending. Thus, to estimate the β parameter in accordance with Eq. (4.1), the α_i parameters from testing unblended tank wastes were assigned from corresponding test temperatures (Fiskum et al. 2021b). The Cs CST capacity (α_i) for AW-102 tank waste was calculated from data reported by Rovira et al. (2019). The assigned α_i and calculated β parameters for the various curve fits are summarized in Table 4.1.

Table 4.1. Summary of Freundlich/Langmuir Hybrid Equilibrium Model α_i and β Parameters

Process Temp. (°C)	Matrix ^(a)	α_i , (mmoles Cs/g CST)	β , (Cs M)	Reference
15.9	AP-107 tank waste	0.782	5.53E-4	Fiskum et al. 2021b
21.0		0.817	6.45E-4	Fiskum et al. 2021b
15.9	AP-105 tank waste	0.475	4.05E-4	Fiskum et al. 2021b
21.0		0.510	4.75E-4	Fiskum et al. 2021b
26.0	AW-102 tank waste	0.730	7.14E-4	^(b)
15.7	Blend B1	0.475	3.26E-4	Current testing
	Blend B2	0.475	3.33E-4	
	Blend B3	0.475	3.24E-4	
24.3	Blend B1	0.510	5.55E-4	
	Blend B2	0.510	5.61E-4	
	Blend B3	0.510	5.37E-4	
15.7	Blend C1	0.730	4.52E-4	
	Blend C2	0.730	4.45E-4	
	Blend C3	0.730	4.89E-4	
24.3	Blend C1	0.730	7.16E-4	
	Blend C2	0.730	6.55E-4	
	Blend C3	0.730	6.67E-4	
15.7	Blend D1	0.475	3.29E-4	
	Blend D2	0.475	3.27E-4	
	Blend D3	0.475	3.09E-4	
24.3	Blend D1	0.510	5.10E-4	
	Blend D2	0.510	5.30E-4	
	Blend D3	0.510	4.99E-4	
13.4	AW-102 tank waste	0.730	4.27E-4	
15.7		0.730	4.69E-4	
24.3		0.730	6.82E-4	

(a) See Table 3.3 for identification of tank waste blends.

(b) The α_i and β parameters were calculated from data reported by Rovira et al. 2019.

Table 4.2 summarizes the equilibrium constant, K_d (mL/g), and Q values for the various tank wastes and blends at the feed Cs concentrations. The relative standard deviation for each tank waste blend set at a given temperature was <6%. This tight precision indicated minimal impact on Cs uptake with changes in blend ratios. The DSA limit threshold (141,600 Ci ¹³⁷Cs per ion exchange column; Anderson 2020) was reevaluated specifically for the AP-105 (Blend B3) and AW-102 (Blend C2) Cs concentrations and ¹³⁷Cs mass fractionations. The maximum Cs loading is 0.108 mmoles/g for AP-105 and 0.107 mmoles/g for AW-102; the maximum K_d value is 1860 mL/g for AP-105 and 2300 mL/g for AW-102. There were no cases where Q and K_d values exceeded the DSA threshold limits for TSCR processing.

Table 4.2. Summary of K_d and Q Values at Equilibrium Cs Concentration

Process Temp. (°C)	Matrix ^(a)	K_d (mL/g) ^(b)	Q (mmoles Cs/g CST)	RPD
15.9	AP-107 tank waste	1255 ^(c)	0.0868 ^(c)	NA
21.0	6.91×10 ⁻⁵ M Cs	1143 ^(c)	0.0790 ^(c)	NA
15.9	AP-105 tank waste	1029 ^(c)	0.0582 ^(c)	NA
21.0	5.65×10 ⁻⁵ M Cs	960 ^(c)	0.0543 ^(c)	NA
26.0	AW-102 tank waste 4.58×10 ⁻⁵ M Cs	961 ^(d)	0.0440 ^(d)	NA
15.7	Blend B1	1241	0.0702	2.1
	Blend B2	1218	0.0692	
	Blend B3	1242	0.0721	
24.3	Blend B1	835	0.0472	3.3
	Blend B2	825	0.0469	
	Blend B3	857	0.0498	
15.7	Blend C1	1464	0.0677	2.9
	Blend C2	1484	0.0690	
	Blend C3	1360	0.0652	
24.3	Blend C1	957	0.0443	5.4
	Blend C2	1040	0.0484	
	Blend C3	1021	0.0490	
15.7	Blend D1	1236	0.0689	1.4
	Blend D2	1242	0.0688	
	Blend D3	1307	0.0706	
24.3	Blend D1	901	0.0502	2.0
	Blend D2	871	0.0483	
	Blend D3	922	0.0498	
13.4	AW-102 tank waste 4.58×10 ⁻⁵ M Cs	1542	0.0706	NA
15.7		1418	0.0649	NA
24.3		1003	0.0459	NA

(a) See Table 3.5 for the blended tank waste Cs concentrations.

(b) K_d calculated as follows: Q, mmoles/g, divided by equilibrium Cs concentration, mmoles/mL.

(c) Fiskum et al. 2021b.

(d) The K_d and Q parameters were calculated from data reported by Rovira et al. 2019.

NA = not applicable

Figure 4.5 shows the Q vs. temperature curve for AW-102 tank waste following the bulk ion exchange in the hot cell to remove Cs/¹³⁷Cs. The slope was similar to those previously reported (Fiskum et al. 2021b). Given the trendline, process temperatures would need to fall to ~1.4 °C for this waste to exceed the DSA limit; however, salts would precipitate before this temperature is reached. For comparison, the AW-102 batch contact test result from hot cell testing (Rovira et al. 2019) is included in Figure 4.5; this result was obtained from contacting neat AW-102 (i.e., not previously processed to remove Cs). This result was 5% higher than the trendline established from the post-Cs removed AW-102 sample. This deviation may be attributed to slight modification of the matrix competitor cations as a result of processing through the CST columns; however, the deviation was generally within the expected experimental uncertainty (estimated at ±10%). Figure 4.5 also compares the blended AW-102 tank wastes (Blends C1 and C3) with the neat AW-102 tank waste. The results agreed within 7% of the trendline.

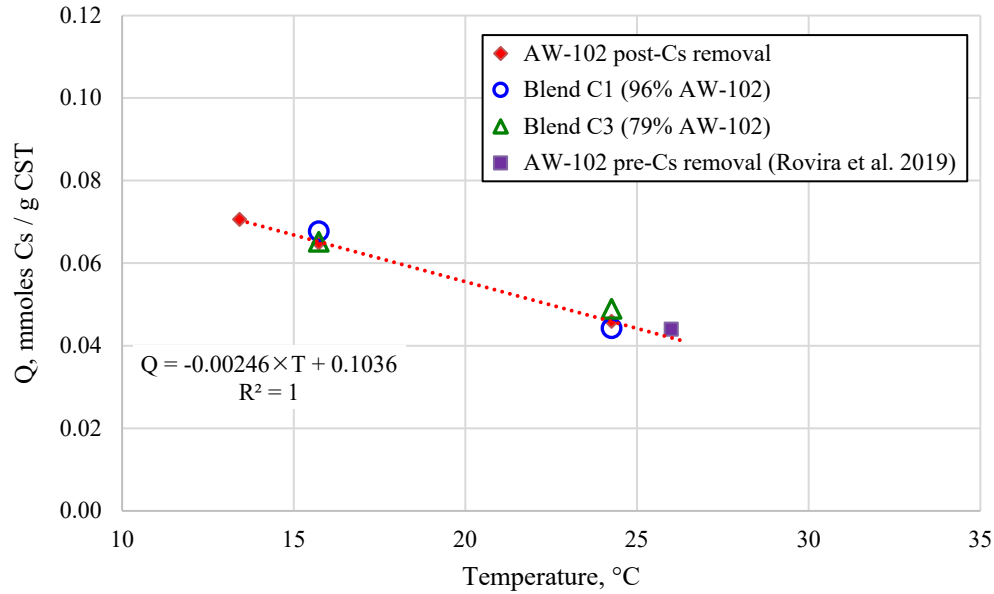


Figure 4.5. Q Dependence on Temperature for AW-102 Tank Waste Post Hot Cell Cs Removal

Figure 4.6 and Figure 4.7 provide similar assessments for AP-105 tank waste (neat) and the AP-105 tank waste blended with AP-107 and AW-102, respectively. The results for the blended wastes were compared to the trendline established by neat AP-105. The results from the tests conducted at 15.7 °C deviated +10% to +15% higher than expected, whereas results from tests conducted at 24.3 °C were within 7% of the expected Q value. The precision of the test results was tight, within 5%; therefore, the higher deviation from the trend line at 15.7 °C appeared to be significant (not associated with experimental uncertainty).

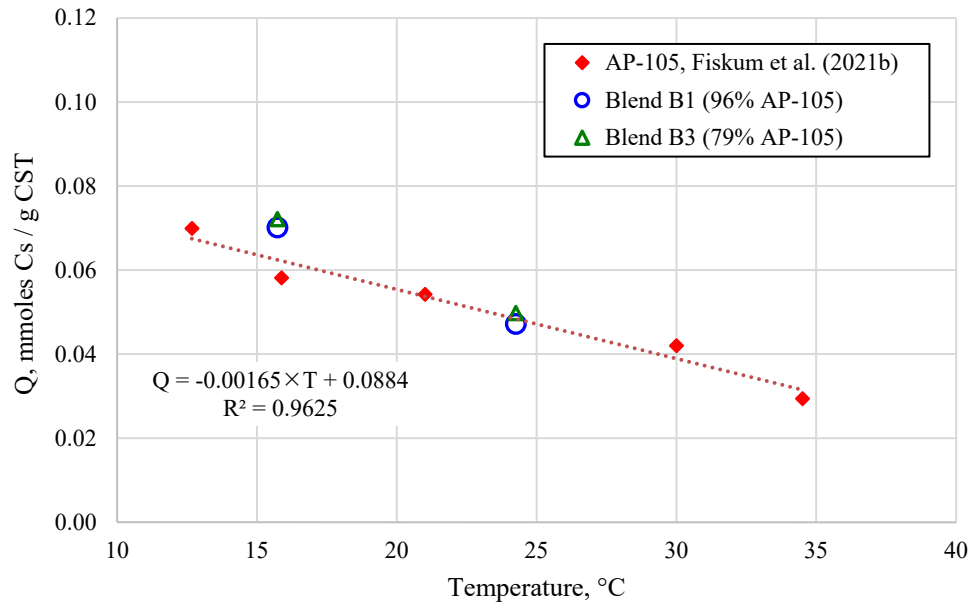


Figure 4.6. Q Dependence on Temperature, Blends B1 and B3 with AP-105 and AP-107 Tank Wastes, Post Hot Cell Cs Removal

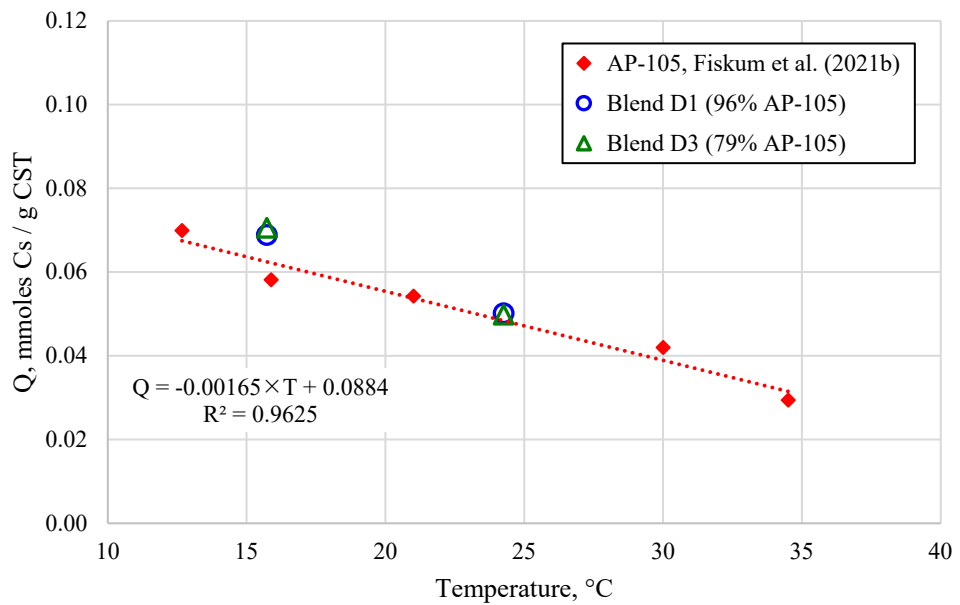


Figure 4.7. Q Dependence on Temperature, Blends D1 and D3 with AP-105 and AW-102 Tank Wastes, Post Hot Cell Cs Removal

5.0 Conclusions

Cesium isotherms were developed for blends of three tank wastes: AP-105/AP-107, AP-105/AW-102, and AW-102/AP-105, each at three nominal blend volume ratios: 21.8:1, 12.9:1, and 3.8:1. Each blend ratio was contacted with CST at 15.7 and 24.3 °C. Additionally, AW-102 tank waste was batch-contacted at three process temperatures: 13.4, 15.7, and 24.3 °C. All tank wastes had been previously processed to remove Cs/¹³⁷Cs such that batch contact testing could be conducted with contact handling. Only the linear portion of the Freundlich/Langmuir Hybrid equilibrium model (Hamm et al. 2002) was evaluated by adding stable Cs to attain 1.30×10^{-4} , 3.40×10^{-4} , and 8.75×10^{-4} M Cs. This allowed for Q (mmoles Cs/g CST) to be evaluated at the equilibrium feed Cs concentration. Batch contact testing was conducted with CST lot 2002009604 in nominal contact phase ratios of 200 (0.075 g dry CST per 15 mL of solution). Contact times lasted nominally 238 h at each temperature.

The following conclusions were made from this testing:

- Blending AW-102 with AP-105, and AP-105 with AP-107, did not result in visually discernable precipitation or separable phases.
- The K_d and Q values were determined at the best-estimate Cs concentration feed conditions (reference date June 21, 2021). The K_d and Q values did not exceed the maximum TSCR column loading of 141,600 Ci (¹³⁷Cs) at either 15.7 or 24.3 °C contact temperature.
- The different blend ratios did not appreciably alter the Q values; the relative standard deviations for the sets of three blend ratios (~3.8 to ~22) at a given temperature were within 6%.
- None of the AW-102 tank waste tests processed at 13.4, 15.7, and 24.3 °C exceeded 0.10 mmoles Cs loading per gram CST and are thus expected to meet the DSA limit of 141,600 Ci ¹³⁷Cs per column. The maximum Q value was 0.0706 mmoles Cs/g CST at 13.4 °C at the nominal equilibrium Cs feed concentration of 4.58×10^{-5} M Cs.
- A linear relationship for Q versus temperature was established for AW-102 tank waste, where Q decreased as temperature increased.
- The blended tank waste Q values did not appreciably differ from the Q vs. temperature relationship established for unblended tank wastes AP-105 and AW-102. Most blended tank wastes values were within 10% of the calculated/expected values based on the curve fit; higher deviations (up to +15%) were found at the lower test temperature of 15.7 °C.

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Appendix A – Batch Contact Data

Batch contact raw data are provided in this appendix, inclusive of F-factor determined at 105 and 427 °C, crystalline silicotitanate (CST) dry masses, tank waste contact volumes, initial and equilibrium Cs concentrations, and corresponding K_d and Q values. The dry CST masses were based on the 105 °C drying temperature. Using the higher 427 °C drying temperature decreased the F-factor ~10%. Thus, applying the 427 °C F-factor would increase K_d and Q values by ~10%.

Table A.1 provides the experimental results associated with tank waste blends of AP-105 and AP-107 (see Table 3.3 in the body of this report for blend ratios associated with the blended tank waste identifications). These data were used to produce the isotherms at two contact temperatures (Figure 4.1 in the body of this report).

Table A.1. Tank Wastes Blends B1, B2, and B3 Isotherm Data at 15.7 and 24.3 °C

Blend ID	Sample ID	Dry CST Mass ^(a) (g)	Tank Waste Vol. (mL)	Initial Cs Conc. (M)	Equil. Cs Conc. (M)	K_d (mL/g)	Q (mmoles Cs/g)
15.7 °C							
B1	TI119B1-S4-16	0.0751	14.308	1.33E-4	1.78E-5	1238	2.20E-2
B1	TI119B1-S5-16	0.0745	14.896	3.41E-4	4.48E-5	1309	5.92E-2
B1	TI119B1-S6-16	0.0763	14.756	8.90E-4	1.18E-4	1275	1.49E-1
B2	TI119B2-S4-16	0.0756	14.660	1.29E-4	1.81E-5	1189	2.15E-2
B2	TI119B2-S5-16	0.0752	14.481	3.50E-4	4.42E-5	1327	5.88E-2
B2	TI119B2-S6-16	0.0750	15.465	8.79E-4	1.24E-4	1249	1.55E-1
B3	TI119B3-S4-16	0.0749	14.295	1.29E-4	1.69E-5	1271	2.15E-2
B3	TI119B3-S5-16	0.0761	15.371	3.36E-4	4.46E-5	1318	5.90E-2
B3	TI119B3-S6-16	0.0780	15.506	8.76E-4	1.24E-4	1196	1.49E-1
24.3 °C							
B1	TI119B1-S4-21	0.0749	15.208	1.33E-4	2.84E-5	743	2.13E-2
B1	TI119B1-S5-21	0.0749	15.148	3.41E-4	6.18E-5	912	5.64E-2
B1	TI119B1-S6-21	0.0749	15.175	8.90E-4	1.72E-4	849	1.46E-1
B2	TI119B2-S4-21	0.0750	15.172	1.29E-4	2.68E-5	761	2.06E-2
B2	TI119B2-S5-21	0.0762	15.079	3.50E-4	6.62E-5	855	5.61E-2
B2	TI119B2-S6-21	0.0754	14.762	8.79E-4	1.68E-4	832	1.39E-1
B3	TI119B3-S4-21	0.0753	15.224	1.29E-4	2.65E-5	784	2.08E-2
B3	TI119B3-S5-21	0.0745	15.203	3.36E-4	6.38E-5	868	5.56E-2
B3	TI119B3-S6-21	0.0752	14.860	8.76E-4	1.46E-4	988	1.44E-1

(a) Based on 105 °C F-factor = 0.9310 (427 °C F-factor = 0.8356).

Table A.2 provides the experimental results associated with tank waste blends of AW-102 and AP-105 (see Table 3.3 in the body of this report for blend ratios associated with the blended tank waste identifications). These data were used to produce the isotherms at two contact temperatures (Figure 4.2 in the body of this report).

Table A.2. Tank Wastes Blends C1, C2, and C3 Isotherm Data at 15.7 and 24.3 °C

Blend ID	Sample ID	Dry CST Mass ^(a) (g)	Tank Waste Vol. (mL)	Initial Cs Conc. (M)	Equil. Cs Conc. (M)	K _d (mL/g)	Q (mmoles Cs/g)
15.7 °C							
C1	TI119C1-S4-16	0.0751	15.056	1.31E-4	1.60E-5	1438	2.30E-02
C1	TI119C1-S5-16	0.0762	15.237	3.38E-4	3.83E-5	1558	5.99E-02
C1	TI119C1-S6-16	0.0754	15.156	8.82E-4	1.13E-4	1372	1.55E-01
C2	TI119C2-S4-16	0.0779	15.199	1.28E-4	1.57E-5	1395	2.19E-02
C2	TI119C2-S5-16	0.0764	14.873	3.42E-4	3.61E-5	1640	5.96E-02
C2	TI119C2-S6-16	0.0742	15.799	8.67E-4	1.10E-4	1467	1.61E-01
C3	TI119C3-S4-16	0.0742	14.970	1.31E-4	1.73E-5	1320	2.29E-02
C3	TI119C3-S5-16	0.0747	14.650	3.41E-4	4.17E-5	1405	5.86E-02
C3	TI119C3-S6-16	0.0766	14.824	8.89E-4	1.11E-4	1353	1.50E-01
24.3 °C							
C1	TI119C1-S4-21	0.0776	15.034	1.31E-4	2.31E-5	903	2.09E-2
C1	TI119C1-S5-21	0.0775	15.028	3.38E-4	5.75E-5	940	5.44E-2
C1	TI119C1-S6-21	0.0741	14.968	8.82E-4	1.52E-4	973	1.48E-1
C2	TI119C2-S4-21	0.0767	15.263	1.28E-4	2.06E-5	1040	2.14E-2
C2	TI119C2-S5-21	0.0750	15.237	3.42E-4	5.75E-5	1001	5.78E-2
C2	TI119C2-S6-21	0.0767	15.212	8.67E-4	1.46E-4	981	1.43E-1
C3	TI119C3-S4-21	0.0749	14.826	1.31E-4	2.13E-5	1018	2.17E-2
C3	TI119C3-S5-21	0.0787	15.165	3.41E-4	5.40E-5	1013	5.52E-2
C3	TI119C3-S6-21	0.0766	15.135	8.89E-4	1.57E-4	930	1.45E-1

(a) Based on 105 °C F-factor = 0.9310 (427 °C F-factor = 0.8356).

Table A.3 provides the experimental results associated with tank waste blends of AP-105 and AW-102 (see Table 3.3 in the body of this report for blend ratios associated with the blended tank waste identifications). These data were used to produce the isotherms at two contact temperatures (Figure 4.3 in the body of this report).

Table A.3. Tank Wastes Blends D1, D2, and D3 Isotherm Data at 15.7 and 24.3 °C

Blend ID	Sample ID	Dry CST Mass ^(a) (g)	Tank Waste Vol. (mL)	Initial Cs Conc. (M)	Equil. Cs Conc. (M)	K _d (mL/g)	Q (mmoles Cs/g)
15.7 °C							
D1	TI119D1-S4-16	0.0782	15.167	1.28E-4	1.76E-5	1212	2.14E-2
D1	TI119D1-S5-16	0.0761	15.299	3.35E-4	4.56E-5	1288	5.83E-2
D1	TI119D1-S6-16	0.0743	15.975	8.49E-4	1.15E-4	1371	1.58E-1
D2	TI119D2-S4-16	0.0760	14.714	1.29E-4	1.74E-5	1239	2.16E-2
D2	TI119D2-S5-16	0.0782	15.562	3.34E-4	4.46E-5	1288	5.76E-2
D2	TI119D2-S6-16	0.0753	15.797	8.59E-4	1.19E-4	1300	1.55E-1
D3	TI119D3-S4-16	0.0763	15.151	1.29E-4	1.67E-5	1336	2.23E-2
D3	TI119D3-S5-16	0.0749	14.386	3.44E-4	4.31E-5	1331	5.78E-2
D3	TI119D3-S6-16	0.0751	15.157	8.74E-4	1.14E-4	1343	1.53E-1
24.3 °C							
D1	TI119D1-S4-21	0.0772	15.150	1.28E-4	2.41E-5	846	2.04E-2
D1	TI119D1-S5-21	0.0746	15.205	3.35E-4	5.98E-5	938	5.62E-2
D1	TI119D1-S6-21	0.0762	15.180	8.49E-4	1.54E-4	903	1.39E-1
D2	TI119D2-S4-21	0.0758	15.222	1.29E-4	2.55E-5	812	2.08E-2
D2	TI119D2-S5-21	0.0755	15.202	3.34E-4	6.25E-5	875	5.46E-2
D2	TI119D2-S6-21	0.0750	15.036	8.59E-4	1.51E-4	932	1.42E-1
D3	TI119D3-S4-21	0.0750	15.196	1.29E-4	2.40E-5	896	2.13E-2
D3	TI119D3-S5-21	0.0762	15.213	3.44E-4	6.10E-5	924	5.64E-2
D3	TI119D3-S6-21	0.0761	15.167	8.74E-4	1.59E-4	894	1.42E-1

(a) Based on 105 °C F-factor = 0.9310 (427 °C F-factor = 0.8356).

Table A.4 provides the experimental results associated with unblended tank waste AW-102. These data were used to produce the isotherms at three contact temperatures (Figure 4.4 in the body of this report). Note that only the 13.4 °C test set was measured in duplicate (“d” suffix in Sample ID).

Table A.4. AW-102 Tank Waste Isotherm Data at 13.4, 15.7, and 24.3 °C

Sample ID	Dry CST Mass ^(a) (g)	AW-102 Vol. (mL)	Initial Cs Conc. (M)	Equil. Cs Conc. (M)	K _d (mL/g)	Q (mmoles Cs/g)
13.4 °C						
TI119E-S4-13	0.0768	14.747	1.30E-4	1.47E-5	1520	2.22E-2
TI119E-S5-13	0.0745	14.467	3.42E-4	3.59E-5	1646	5.95E-2
TI119E-S6-13	0.0776	14.492	8.87E-4	9.82E-5	1502	1.47E-1
TI119E-S4-13d	0.0749	15.416	1.30E-4	1.48E-5	1605	2.38E-2
TI119E-S5-13d	0.0759	16.128	3.42E-4	3.99E-5	1606	6.42E-2
TI119E-S6-13d	0.0754	16.538	8.87E-4	1.26E-4	1316	1.67E-1
15.7 °C						
TI119E-S4-16	0.0762	15.206	1.30E-4	1.64E-5	1396	2.28E-2
TI119E-S5-16	0.0762	15.200	3.42E-4	4.07E-5	1472	6.02E-2
TI119E-S6-16	0.0774	15.146	8.87E-4	1.11E-4	1367	1.52E-1
24.3 °C						
TI119E-S4-21	0.0787	15.186	1.30E-4	2.15E-5	981	2.10E-2
TI119E-S5-21	0.0762	15.177	3.42E-4	5.87E-5	965	5.65E-2
TI119E-S6-21	0.0742	15.197	8.87E-4	1.52E-4	996	1.51E-1

(a) Based on 105 °C F-factor = 0.9310 (427 °C F-factor = 0.8356).

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