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Characterization Results for the 2020 Tank Farm 2H and 3H Evaporator Overhead Samples

S. C. Lucatero

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REVIEWS AND APPROVALS

AUTHOR:

____

S. C. Lucatero, Separation Sciences & Engineering

TECHNICAL REVIEWER:

____

M. S. Hay, Separation Sciences & Engineering

APPROVALS:

____

B. J. Wiedenman, Manager, Separation Sciences & Engineering

____

S. D. Fink, Director, Chemical Processing Sciences

____

P. W. Norris, Manager, Tank Farm Facility Engineering - ETP

EXECUTIVE SUMMARY

The 2020 annual 2H and 3H Evaporator overhead samples were received at Savannah River National Laboratory (SRNL) on December 22, 2020. The delivered samples were analyzed for concentration of cesium-137 (^{137}Cs), strontium-90 (^{90}Sr) and iodine-129 (^{129}I) to validate compliance with the Effluent Treatment Project Waste Acceptance Criteria (ETP WAC, Rev. 7).[1] The concentration of all three radionuclides, ^{137}Cs, ^{90}Sr, and ^{129}I, in the 2H Evaporator overhead sample were found to be less than the corresponding ETP WAC limits. Radiochemical analysis of the 3H Evaporator overhead sample indicated a ^{137}Cs content about an order of magnitude higher than the acceptable threshold value, and ^{90}Sr and ^{129}I concentrations within the acceptance limits.

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LIST OF ABBREVIATIONS

ARD	Analytical Research and Development
ELN	Electronic Laboratory Notebook
ETP	Effluent Treatment Project
%RSD	percent relative standard deviation
SRNL	Savannah River National Laboratory
WAC	Waste Acceptance Criteria
WCP	Waste Compliance Plan

1.0 INTRODUCTION

The purpose of the H-Tank Farm evaporator system is to reduce the amount of liquid volume (e.g., water) in high-level radioactive waste by means of a boiling process. The vapor phase in the overhead space of evaporators is circulated through a condensation unit and the generated liquid streams are sent to ETP for further decontamination and eventual release. The condensed overhead streams are analyzed for contaminant content, and species exceeding ETP WAC limits[1] are returned to the feed tank and recycled back to the evaporator, as directed per the Waste Compliance Plan (WCP).[2] In fulfilling such regulatory requirements, annual samples of the 2H and 3H Evaporator overhead streams were submitted to SRNL on December 22, 2020, for radiochemical characterization. The concentrations of cesium-137 (^{137}Cs), strontium-90 (^{90}Sr) and iodine-129 (^{129}I) were measured in the as received samples.

2.0 EXPERIMENTAL PROCEDURE

The 2H and 3H annual evaporator overhead samples were delivered at E-wing of SRNL on December 22, 2020 in disposable plastic containers. Both samples looked visually clear, although the 3H sample exhibited a slightly stronger yellowish color than the 2H sample, which showed a paler tone. The total volume of each sample was approximately 200 mL. Owing to the relatively low radioactivity of the as received samples, undiluted aliquots were submitted to Analytical Research and Development (ARD) in green shielded bottles for pertinent radiochemical analyses. De-ionized water blank samples were processed in parallel for Quality Assurance purposes and to evaluate potential sample contamination.

The characterization techniques for duplicate analysis of ^{137}Cs, ^{90}Sr and ^{129}I applied are specified in the Task Technical and Quality Assurance Plan for the Annual Overheads Sample Analysis for 2H/3H Evaporators.[3] The ^{137}Cs concentration in the samples was measured via gamma spectrometry, whereas the ^{90}Sr (^{129}I) content was determined by first undertaking a radiochemical separation followed by liquid scintillation counting (low energy gamma photon spectroscopy).

The density of as received 2H/3H Evaporator overhead samples was determined at 25ºC using 2 mL graduated glass vials, which were tare weighed using an analytical balance. Each vial was filled with sample to the 2 mL mark, the weight was then determined by difference and recorded for density calculations. These steps were repeated three times and average values along with percent relative standard deviation (%RSD) calculated afterwards. The density of de-ionized water was also measured using the same procedure.

As per waste disposal requirements for the High Activity Drain and Quality Assurance assessment purposes, the total mercury and anion concentrations were analyzed in the 2H and 3H samples by direct mercury analysis and ion chromatography for anions. Appendix A provides a summary of these results.

3.0 RESULTS AND DISCUSSION

A summary of the analytical results along with the applicable ETP WAC limits for ^{137}Cs, ^{90}Sr and ^{129}I is provided in Table 3-1. The average concentration values listed therein were calculated from duplicate determinations performed by ARD. The percent relative standard deviation (%RSD, defined as [standard deviation / average] *100) was calculated only for values that were above the detection limits. All three radionuclide concentrations in the 2H Evaporator overhead sample were found to be less than the corresponding ETP WAC limits. However, the 3H Evaporator overhead sample exhibited a ^{137}Cs content equal to 4.61E+03 dpm/mL (5.0% uncertainty), which exceeded the ETP WAC threshold by approximately an order of magnitude. In addition, there is WAC deviation documentation that has moved the point of compliance and limits for ^{137}Cs,[8] the measured value also exceeded the deviation limit. The facility has a process that procedurally requires recycling overheads back to the feed tank if the results exceed the WAC. Since it was the case with the concentration of ^{137}Cs in the 3H Evaporator overhead sample, the facility did confirm the transfer of the overheads tank contents to the feed tank. The concentration of ^{90}Sr and ^{129}I in the 3H Evaporator overhead sample fell below the acceptance limits; duplicate analysis indicated a ^{90}Sr content above and another below the detection levels; hence, the average value listed in Table 3-1 is preceded by a *less than or equal* sign.

The average ^{137}Cs concentration in the 2H Evaporator overhead sample was approximately half of the ETP WAC value, 1.58E+02 dpm/mL, and was slightly higher than corresponding values reported in recent annual characterizations: 7.04E+01,[4] 6.97E+01[5] and 9.13E+01[6] dpm/mL. Analytical determinations of both the 2H and 3H Evaporator overhead samples indicated that ^{90}Sr and ^{129}I were so low in concentration that they fell below the limits of detection and are therefore reported as values preceded by the *less than* symbol. Previous annual characterizations[5-6] have similarly reported ^{90}Sr and ^{129}I contents below the detection limits in 2H Evaporator overhead samples. Table 3-1 also includes radionuclide concentrations measured in de-ionized water blank samples, which all, ^{137}Cs, ^{90}Sr and ^{129}I, were found to be less than the minimum detectable concentrations.

Table 3-1 Results of radiochemical analysis of 2H and 3H Evaporator overhead samples.

Analyte	2H Evaporator overhead sample (dpm/mL) (%RSD)	3H Evaporator overhead sample (dpm/mL) (%RSD)	De-ionized water sample (dpm/mL)	ETP WAC limits (dpm/mL)[7]
^{137}Cs	1.58E+02 (0.45)	4.61E+03(0.00)[a]	<2.64E+00[b]	3.28E+02 (1.3E+03[8])
^{90}Sr	<9.49E+00[b]	≤8.43E+00[b]	<7.86E+00[b]	1.76E+02
^{129}I	<2.32E-01[b]	<1.59E-01[b]	<2.09E-01[b]	1.00E+00

[a]Results of duplicate analysis were identical. [b]%RSD not applicable.

The density of the 2H and 3H Evaporator overhead samples was closely comparable, 0.992 (%RSD = 0.23%) and 0.997 gr/mL (%RSD = 0.15%), respectively, whereas the measured density of de-ionized water was 1.002 g/mL (%RSD = 0.20%).

3.1 Quality Assurance

This report was developed in accordance with the protocols identified in Task Technical and Quality Assurance Plan SRNL-RP-2021-00045.[3] Requirements for performing reviews of technical reports and the extent of review are established in manual the E7 Manual, Procedure 2.60. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2. This review and all the work meets or exceeds the acceptable criteria to comply with the Technical Task Request[7] requesting this work with a functional classification of Production Support. The data from the experiments entailed herein is contained in an Electronic Laboratory Notebook (ELN).[9]

4.0 CONCLUSIONS

Characterization results of the 2020 annual overhead samples for 2H and 3H Evaporators summarized in this report compare the radionuclide contents between the as received samples and the ETP WAC limits. The concentration of ^{137}Cs, ^{90}Sr, and ^{129}I in the 2H Evaporator overhead sample exhibited compliance with the ETP WAC. Radiochemical analysis of the 3H Evaporator overhead sample indicated a ^{137}Cs content about an order of magnitude higher than the acceptable threshold value, and concentrations of ^{90}Sr and ^{129}I within the acceptance limits. The radionuclide concentrations measured in these samples were comparable to those reported in previous annual characterizations, except the ^{137}Cs content of the 3H sample, which exceeded the ETP WAC limit.

5.0 REFERENCES

1. "F/H Effluent Treatment Facility Waste Acceptance Criteria," X-SD-H-00009, Revision 7, December 16, 2020.
2. S. Campbell, "F/H Tank Farms Waste Compliance Plan for Transfers to the Effluent Treatment Project," X-WCP-H-00013, Revision 8, June 8, 2016.
3. S. C. Lucatero, "Task Technical and Quality Assurance Plan for the Annual Overheads Sample Analysis for 2H/3H Evaporators," SRNL-RP-2021-00045, Rev. 0, February 2021.
4. Nicholson, J. C., "Characterization Results for the March 2016 H-Tank Farm 2H Evaporator Overhead Samples," SRNL-STI-2016-00253, Rev. 1, September 2016.
5. T. T. Truong, "Characterization Results for the January 2017 H-Tank Farm 2H Evaporator Overhead Sample", SRNL-STI-2017-00166, Rev. 0, April 10, 2017.
6. A. L. Washington, "Characterization Results for the July 2018 H-Tank Farm 2H Evaporator Overhead Samples," SRNL-STI-2019-00069, February 2019.
7. P. W. Norris, "2020 Annual Overhead Samples Analysis for 2H/3H Evaporator Overhead Samples," X-TTR-H-00115, Rev. 0, 12/22/2020.
8. "Request for Deviation to ETP Waste Acceptance Criteria," X-WDEV-H-00010, Revision 0, December 9, 2020.
9. S. C. Lucatero, Radiochemical Analysis of 2H/3H Evaporator Overheads, K7753-00481-01, SRNL E-Notebook (Production), Savannah River National Laboratory, February 2021.

Appendix A. Total mercury and anion determinations of 2H and 3H Evaporator overhead samples.

	2H Evaporator overhead sample (one sigma % uncertainty)	3H Evaporator overhead sample (one sigma % uncertainty)
Total mercury, mg/L	1.867 (10)	12.742 (10)
Fluoride, F^{-1}, µg/mL	<5 (10)	<5 (10)
Formate, CHO_2^{-1}, µg/mL	<5 (10)	<5 (10)
Chloride, Cl^{-1}, µg/mL	<5 (10)	<5 (10)
Nitrite, NO_2^{-1}, µg/mL	<5 (10)	<5 (10)
Nitrate, NO_3^{-1}, µg/mL	<5 (10)	<5 (10)
Phosphate, PO_4^{-3}, µg/mL	<5 (10)	<5 (10)
Sulfate, SO_4^{-2}, µg/mL	<5 (10)	<5 (10)
oxalate, $C_2O_4^{-2}$, µg/mL	<5 (10)	<5 (10)
Bromide, Br^{-1}, µg/mL	<50 (10)	<50 (10)

DISTRIBUTION LIST

cj.bannochie@srnl.doe.gov
alex.cozzi@srnl.doe.gov
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connie.herman@srnl.doe.gov
dennis.jackson@srnl.doe.gov
brady.lee@srnl.doe.gov
joseph.manna@srnl.doe.gov
daniel.mccabe@srnl.doe.gov
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azadeh.samadi-dezfouli@srs.gov
christie.sudduth@srs.gov
vijay.jain@srs.gov
arthur.wiggins@srs.gov
Records Administration (EDWS)