Contract No:

This document was prepared in conjunction with work accomplished under Contract No. 89303321CEM000080 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

Disclaimer:

This work was prepared under an agreement with and funded by the U.S. Government. Neither the U.S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied:

- 1) warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or
- 2) representation that such use or results of such use would not infringe privately owned rights; or
- endorsement or recommendation of any specifically identified commercial product, process, or service.

Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.



A U.S. DEPARTMENT OF ENERGY NATIONAL LAB • SAVANNAH RIVER SITE • AIKEN, SC • USA

Characterization Results for the March 2022 Tank Farm 3H Evaporator Overhead Sample

J. R. Dekarske

L. N. Oji

D. P. DiPrete

June, 2022

SRNL-STI-2022-00212, Revision 0

DISCLAIMER

This work was prepared under an agreement with and funded by the U.S. Government. Neither the U.S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied:

- 1. warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or
- 2. representation that such use or results of such use would not infringe privately owned rights; or
- 3. endorsement or recommendation of any specifically identified commercial product, process, or service.

Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.

Printed in the United States of America

Prepared for U.S. Department of Energy

Keywords: Effluent Treatment Project, WAC, ¹³⁷Cs, ⁹⁰Sr, ¹²⁹I.

Retention: Permanent

Characterization Results for the March 2022 Tank Farm 3H **Evaporator Overhead Sample**

J. R. Dekarske

L. N. Oji

D. P. DiPrete

June, 2022



REVIEWS AND APPROVALS

AUTHORS:	
J. R. Dekarske, Chemical Flowsheet Development	Date
L. N. Oji, Chemical Flowsheet Development	Date
D. P. DiPrete, Environmental and Legacy Management TECHNICAL REVIEW:	Date
M. S. Hay, Chemical Flowsheet Development, Reviewed per E7 2.60	Date
APPROVAL:	Bute
Gregg Morgan, Manager, Chemical Flowsheet Development	Date
Frank Pennebaker, Director, Chemical Processing and Characterization	Date
Phillip Norris, SRMC Evaporator & ETP Engineering, Manager	Date

ACKNOWLEDGEMENTS

The authors acknowledge assistance in the completion of sample transfers and handling tasks by Shirley McCollum and the completion of sample analyses tasks by David DiPrete.

EXECUTIVE SUMMARY

On an annual basis, Savannah River Mission Completion (SRMC) provides 2H and 3H evaporator overhead samples to Savannah River National Lab (SRNL) to be analyzed per Section 5.2 of the Effluent Treatment Project (ETP) Waste Compliance Plan (WCP)ⁱⁱ and the Waste Acceptance Criteria (WAC)ⁱⁱ.

This report presents characterization results for the March 2022 3H evaporator overhead sample. The sample was clear and colorless with no visible solids. The results provide measurements for cesium-137 (¹³⁷Cs), strontium-90 (⁹⁰Sr), and iodine-129 (¹²⁹I) with the radionuclide concentration limits specified by the WAC.

These analyses were performed on a single sample replicate. A summary of the analytical results for the 3H evaporator overhead sample includes the following.

The measured cesium-137 activity in the 3H evaporator overhead sample is 4.59E+02 dpm/mL which is above the ETP WAC limit of 3.28E+02 dpm/mL.

The strontium-90 activity in the 3H evaporator overhead sample is <2.95E+01 dpm/mL. This is below the ETP WAC limit of 1.76E+02 dpm/mL.

The iodine-129 activity in the 3H evaporator overhead sample is <1.92E-02 dpm/mL. This is below the ETP WAC limit of 1.00E+00 dpm/mL.

TABLE OF CONTENTS

LIST OF TABLES	viii
LIST OF FIGURES	viii
LIST OF ABBREVIATIONS	viii
1.0 Introduction	1
2.0 Experimental Procedure	1
3.0 Results and Discussion	2
4.0 Conclusions	4
5.0 Quality Assurance	5
6.0 References	5

LIST OF TABLES

Table 3-1. Results for 2022 3H Evaporator Overhead and Blank samples: ¹³⁷ Cs, ¹²⁹ I, and ⁹⁰ Sr (March 2022).
Table 3-2. Historical Analytical Results for 3H Evaporator Overhead Samples: ¹³⁷ Cs, ¹²⁹ I, and ⁹⁰ Sr4
LIST OF FIGURES
Figure 3-1. Photograph of the 3H Evaporator Overhead Sample in a Plastic Beaker

LIST OF ABBREVIATIONS

ETF Effluent Treatment Facility
ETP Effluent Treatment Project
HPGe High Purity Germanium

MDA Minimum Detectable Activity

ND Not Detected

SRMC Savannah River Mission Completion
SRNL Savannah River National Laboratory

TTQAP Task Technical and Quality Assurance Plan

TTR Technical Task Request
WAC Waste Acceptance Criteria
WCP Waste Compliance Plan

1.0 Introduction

To minimize and reduce the large amount of high-level liquid waste volume at the Savannah River Site (SRS), the 2H and 3H evaporators were constructed and began operations in H Area in 1982 and 2000, respectively. The evaporation process is performed through boiling the liquid waste in the evaporator cell, cooling and condensing the overhead vapors in the condenser cell, followed by collecting the condensate in the overheads cell. The low-level liquid waste is further treated at the Effluent Treatment Facility (ETF) prior to release into the environment.

On an annual basis, Savannah River Mission Completion (SRMC) provides 2H and 3H evaporator overhead samples to Savannah River National Laboratory (SRNL) for select radionuclide (137Cs, 90Sr, and 129I) characterizations to ensure that the Effluent Treatment Project (ETP) Waste Acceptance Criteria (WAC) for these radionuclides are met as specified in Section 5.2 of the ETP Waste Compliance Plan (WCP)ⁱ and the WACⁱⁱ. In this report, following the specified Technical Task Request (TTR)ⁱⁱⁱ and Task Technical and Quality Assurance Plan (TTQAP)^{iv}, the March 2022 3H evaporator overhead sample was analyzed for cesium-137 (137Cs), strontium-90 (90Sr), and iodine-129 (129I) with the radionuclide concentration limits specified by the WAC.

2.0 Experimental Procedure

The 3H evaporator overhead sample was received on March 17, 2022 at SRNL. Since the "as-received" sample radiation dose rate was low [extremity, skin, and whole body were below instrument detection limit (ND)], the container was moved to a radiological hood for inspection. Approximately 200 mL of sample was collected from the receipt vessel and was transferred into a clear plastic beaker for visual inspection. The 3H evaporator overhead and distilled/deionized water blank samples were submitted for: 1) gamma spectroscopy (¹³⁷Cs) and 2) chemical separations followed by beta counting (¹²⁹I and ⁹⁰Sr).

Cs-137 concentrations were determined by gamma spectrometry. 50 milliliter aliquots of sample were analyzed directly for analysis periods of at least 4 hours. The samples were analyzed by shielded, high purity germanium (HPGe) gamma spectrometers. I-129 and Sr-90 were determined by radiochemistry methods. These analytical methods involved separation techniques that enabled radionuclides that were at low concentrations to be measured more accurately and to determine more reliable and lower detection limits. The techniques and methodology for these separations will be summarized here.

Sr-90 Method: 20 milliliter aliquots of each sample was spiked with a stable Sr carrier, and a stable Ce carrier. The Sr carrier was used for separation yielding purposes and the Ce carrier was used to enhance the separation rates of undesirable isotopes such as Y-90, the lanthanides or the actinides. The spiked sample aliquots were acidified with nitric acid, evaporated to dryness and re-dissolved in 8N nitric acid. The Sr in the samples was then extracted using a commercially available Sr extraction resin. This resin also extracts some of the Pu under the conditions used to extract the Sr. The Pu was washed from the resin using an oxalic acid/nitric acid mixture. The Sr was eluted from the resin, and the resulting solution concentrated. A portion of the purified Sr solution was neutron activated in a Cf-252 neutron activation facility at SRNL to determine the total Sr and in order to calculate the fraction of Sr isolated by the procedure. A second portion of each of the Sr fractions was stored for five to seven days to allow Y-90 to grow in. Each fraction was then counted by liquid scintillation analysis using a Low-Level Perkin Elmer Tri-Carb Liquid scintillation counter to determine the Y-90 activity in a high energy beta window free of interferences from Sr-90 or any residual beta interferences from isotopes such as Cs-137. The Sr-90 beta activity in each case was calculated from the Y-90 activity. The yields of the stable Sr carriers were applied to the Sr-90 beta activity results to determine Sr-90 activities in the original aliquots of the solutions.

I-129 Method: 50 milliliter aliquots of sample were spiked with a known amount of stable KI to act as an iodine tracer/carrier. The samples were acidified with nitric acid. The samples were decontaminated with a

resin treatment to enhance removal of the actinide elements. The iodine in the sample was then reduced to iodide. The solution was then treated with AgNO₃ in order to precipitate the iodide ion as AgI. The precipitate was analyzed by low energy photon spectrometry to determine the amount of I-129 present. I-129 is detected by its characteristic gamma and x-ray emissions. The precipitate was then neutron activated in a Cf-252 neutron source at SRNL to determine the total amount of iodine present in order to calculate the recovery of I-129 in the radiochemical separation.

3.0 Results and Discussion

A photograph of the "as-received" 3H evaporator overhead sample in a 250 mL capacity plastic container is provided in Figure 3-1. The sample appearance was clear and colorless with no visible solids.

Normally, it is necessary to use a larger sample volume to enhance analyte detection limits and to perform analyses in duplicate, especially in cases where the samples activities for the radionuclides of interest are low. Because the sample volume provided for this March 2022 overhead sample was small, only single analysis results were produced for each analyte instead of duplicates. The analytical results for the characterization of the 3H evaporator overhead and the water blanks are provided in Table 3-1. The ¹³⁷Cs activity in the 3H evaporator overhead is above the ETP WAC limit of 3.28E+02 dpm/mL. This analytical result for ¹³⁷Cs is like the previous year 3H evaporator overhead sample analytical result for ¹³⁷Cs, which was also above the WAC limit. Historical results for 3H evaporator overhead samples are provided in Table 3-2, and a survey of the historical results trends demonstrates that ¹³⁷Cs activity is generally close to the ETP WAC limit except for the 2020 result, which was above the WAC and a factor of ten higher than the others.



Figure 3-1. Photograph of the 3H Evaporator Overhead Sample in a Plastic Beaker

Table 3-1. Results for 2022 3H Evaporator Overhead and Blank samples: ¹³⁷Cs, ¹²⁹I, and ⁹⁰Sr (March 2022).

Analyte	Activity (dpm/mL) (3H Evaporator Overhead)	Activity (pCi/mL) (3H Evaporator Overhead)	Blank Sample (dpm/mL)	ETP WAC limits (dpm/mL) ⁱⁱ
¹³⁷ Cs	4.59E+02 (5.00%) ^β	2.07E+02	<2.78E-01 (MDA)	3.28E+02
⁹⁰ Sr	<2.95E+01 (MDA)	<1.33E+01	<3.73E+01 (MDA)	1.76E+02
129 I	<1.92E-02 (MDA)	<8.65E-03	<4.86E-02 (MDA)	1.00E+00

^β One sigma % uncertainties for all analytical methods.

Table 3-2. Historical Analytical Results for 3H Evaporator Overhead Samples: ¹³⁷Cs, ¹²⁹I, and ⁹⁰Sr.

Analyte	¹³⁷ Cs, dpm/mL	⁹⁰ Sr, dpm/mL	¹²⁹ I, dpm/mL
ETP WAC limits ⁱⁱ	3.28E+02	1.76E+02	1.00E+00
March 2022 3H Evaporator Overhead Sample	4.59E+02	<2.95E+01	<1.92E-02
2020 3H Evaporator Overhead Sample ^v	4.61E+03	≤8.43E+00	<1.59E-01
2015 3H Evaporator Overhead Sample ^{vi}	1.52E+02	<3.47E+00	<4.87E-02
2014 3H Evaporator Overhead Sample ^{vii}	1.52E+02	<1.01E+01	<2.94E-02
2013 3H Evaporator Overhead Sample ^{viii}	2.65E+02	<2.43E+00	<3.33E-02
2011 3H Evaporator Overhead Sample ^{ix}	2.26E+02	<1.64E+01	<6.06E-01

4.0 Conclusions

The March 2022 3H evaporator overhead sample characterization result for ¹³⁷Cs activity (4.59E+02 dpm/mL) was above the ETP WAC limit of 3.28E+02 dpm/mL. The ¹²⁹I activity (<1.92E-02 dpm/mL) and ⁹⁰Sr activity (<2.95E+01 dpm/mL) in the sample were below the ETP WAC limits of 1.00E+00 dpm/mL and 1.76E+02 dpm/mL, respectively.

5.0 Quality Assurance

Requirements for performing reviews of technical reports and the extent of review are established in manual E7 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, a design verification done by document review, meets the acceptance criteria to comply with the TTRⁱⁱⁱ requesting this work with a functional classification of Safety Class and per guidance in the TTQAP^{iv}. Data are recorded in the electronic laboratory notebook system as Experiment ID M0869-00537-06.*

6.0 References

i S. Campbell, "F/H Tank Farms Waste Compliance Plan for Transfers to the Effluent Treatment Project", X-WCP-H-00013, Rev. 8, June 2016.

ii "F/H Effluent Treatment Facility Waste Acceptance Criteria", X-SD-H-00009, Rev. 7, December 2020.

iii Technical Task Request, "2020 Annual Overheads Sample Analysis for 2H/3H Evaporators", X-TTR-H-00115, Rev. 0, December 2020.

iv 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.

^v S.C. Lucatero, "Characterization Results for the 2020 Tank Farm 2H and 3H Evaporator Overhead Samples", SRNL-STI-2021-00076, Rev. 0, 2021.

vi J.C. Nicholson, "Characterization Results for the October 2015 H-Tank Farm 3H Evaporator Overhead Samples", SRNL-STI-2016-00010, Rev. 0, 2016.

vii A.L. Washington, II, "Characterization Results for the 2014 HTF 3H & 2H Evaporator Overhead Samples", SRNL-STI-2015-00198, Rev. 0, 2015.

viii A.L. Washington, II, "Characterization Results for the 2013 HTF 3H Evaporator Overhead Samples", SRNL-STI-2013-00525, Rev. 0, 2013.

^{ix} A.L. Washington, II, "Report on the Analysis of WAC Samples from Evaporator Overheads for 2011", SRNL-STI-2011-00660, Rev. 0, 2011.

x "Technical Reviews," E7 Manual, Procedure 2.60, Rev. 18, 2019.

xi "Savannah River National Laboratory Technical Report Design Check Guidelines", WSRC-IM-2002-00011, Rev.2, 2004.

xii J.R. Dekarske: ELN: M0869-00537-06 (Electronic Notebook (Production)); SRNL, Aiken, SC 29808 (2022).

Distribution:

John.dekarske@srnl.doe.gov Lawrence.oji@srnl.doe.gov David.diprete@srnl.doe.gov Michael.hay@srnl.doe.gov cj.bannochie@srnl.doe.gov alex.cozzi@srnl.doe.gov William.bates@srnl.doe.gov marion.cofer@srnl.doe.gov connie.herman@srnl.doe.gov brady.lee@srnl.doe.gov joseph.manna@srnl.doe.gov gregg.morgan@srnl.doe.gov frank.pennebaker@srnl.doe.gov william.ramsey@srnl.doe.gov Marissa.Reigel@srnl.doe.gov eric.skidmore@srnl.doe.gov michael.stone@srnl.doe.gov william.swift@srnl.doe.gov boyd.wiedenman@srnl.doe.gov jocelyn.lampert@srnl.doe.gov mary.whitehead@srnl.doe.gov jeffrey.crenshaw@srs.gov james.folk@srs.gov curtis.gardner@srs.gov timothy.littleton@srs.gov Anna.Murphy@srs.gov Anthony.Robinson@srs.gov mark-a.smith@srs.gov tony.polk@srs.gov patricia.suggs@srs.gov thomas.temple@srs.gov david02martin@srs.gov celia.aponte@srs.gov timothy.baughman@srs.gov Azikiwe.hooker@srs.gov ryan.mcnew@srs.gov phillip.norris@srs.gov azadeh.samadi-dezfouli@srs.gov vijay.jain@srs.gov Bruce.wiersma@srnl.doe.gov arthur.wiggins@srs.gov eric.harrison@srs.gov john.jacobs@srs.gov john.pateracki@srs.gov

Records Administration (EDWS)