Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 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
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.

January 11, 2017

TO: E. N. HOFFMAN

FROM: W. D. KING

Approval: ____
E. N. Hoffman, Manager, EPD/ECPT/SRNL

Technical Review per E7 2.60: ___________________________
M. S. Hay, ACPT/ECPT/SRNL

ANALYSIS RESULTS FOR SUB-SAMPLES OF SRS TANKS 30, 32, AND 39 TO SUPPORT EVALUATIONS OF THE I-129 INVENTORY

Introduction

In order to appropriately model and predict the chemical integrity and performance of cementitious materials used for waste immobilization at the Savannah River Site (SRS), it is critical to understand the I-129 solubility and distribution within the tank farm. Iodine in radioactive waste and in environmental media is typically highly mobile and long lived. Iodine is ubiquitous in SRS tank waste and waste forms. The iodine is assumed to be soluble and present at low levels in Performance Assessments (PAs) for SRS Tank Farms[1] and is one of the dose drivers in the PAs for both the SRS Salt Disposal Facility (SDF) and the H-Area Tank Farm (HTF). Analysis of tank waste samples is critical to understanding the Tank Farm iodine inventory and reducing disposal uncertainty. Higher than expected iodine levels have recently been observed in residual solids isolated from some SRS tanks prior to closure, indicating uncertainty regarding the chemical species involved.[2] If the iodine inventory uncertainty is larger than anticipated, future work may be necessary to reduce the uncertainty. This memorandum satisfies a portion of the work scope identified in Task Plan SRNL-RP-2016-00651.[3] A separate memorandum issued previously, reported historical unpublished I-129 data, a significant portion of which was below detectable analytical limits.[4] This memorandum includes iodine and general chemical analysis results for six archived SRNL samples which were previously reported to have I-129 concentrations below detectable limits. Lower sample dilution factors were used for the current analyses in order to obtain concentrations above detection. The samples analyzed included surface and depth samples from SRS tanks 30, 32, and 39.

Experimental

Six archive samples from SRS Tanks 30, 32, and 39 (specific sample IDs provided in Table 1) were located in the SRNL shielded cells sample inventory and retrieved from storage. After mixing, the density of each supernate sample was measured in duplicate at ambient temperature (17 °C) using ~8.5 mL sub-samples and plastic density tubes which had been pre-calibrated with water to determine the volume contained. Portions of each supernate sample were filtered through separate 0.2 µm polytetrafluoroethylene (PTFE) filter membranes. Filtrate sub-samples were then transferred to shielded sample bottles containing known masses of 5 M HNO_3 or deionized water. Individual samples (no replicates) diluted in acid were analyzed for soluble metals by Inductively Coupled Plasma - Emission Spectroscopy (ICP-ES). Individual samples (no replicates) diluted in water were analyzed for anions by Ion Chromatography (IC) and for hydroxide (Free Hydroxide). Replicate samples diluted in water were analyzed for I-129 using radioanalytical counting methods. Volume-based dilution factors for samples diluted in acid ranged from 9-10. Dilution factors for samples diluted in water ranged from 6.5-7.0.

Quality Assurance

Requirements for performing reviews of technical reports and the extent of review are established in Manual E7, 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. Data are recorded in the Electronic Laboratory Notebook (ELN) system as notebook/experiment number A2341-00117-06.

Results

Major anion and soluble metal concentrations determined by analysis for the six supernate samples are provided in Table 1. I-129 analyses results above detectable limits and consistent with previous results were observed for each sample. Iodine data for surface and depth samples from a given tank were similar except for the Tank 30 samples where the I-129 concentration of the depth sample was 1.7 times greater than the concentration observed for the surface sample. The general chemical composition of each sample was also determined. Sodium concentrations for the samples ranged from 4-12 M with the Tank 32 sample being the most concentrated and the Tank 39 sample being the most dilute. As expected, total sodium concentrations were higher for the depth samples than for the surface samples from each tank, which is indicative of tank stratification. Minimal stratification was observed for the more dilute Tank 39 sample. The sodium concentration for each sample exceeded the sum of the major anions (nitrate, nitrite, and hydroxide) with the balance of the soluble anions likely being dominated by carbonate ion, which was not measured. Trends in the sample densities were consistent with the sodium analysis results.

Table 1. Density and Chemical Analysis Data for Tank 30, 32, and 39 Surface and Depth Samples.

Tank Location	30 Surface	30 Depth	32 Surface	32 Depth	39 Surface	39 Depth
Sample ID	HTF-30-15-69	HTF-30-15-70	HTF-32-15-67	HTF-32-15-68	HTF-39-15-132	HTF-39-15-133
Density (g/mL)	1.371	1.477	1.439	1.495	1.209	1.204
Na (mg/L)	1.98E+05	2.68E+05	2.52E+05	2.72E+05	9.69E+04	1.00E+05
Na (M)	8.62	11.65	10.97	11.85	4.21	4.35
Al (mg/L)	1.28E+04	1.93E+04	1.68E+04	1.82E+04	2.75E+03	3.10E+03
Al (M)	0.47	0.71	0.62	0.67	0.10	0.11
NO_3^- (M)	1.55	1.40	1.88	1.79	1.59	1.73
NO_2^- (M)	1.34	2.06	1.88	2.19	0.27	0.30
OH^- (M)	4.49	6.93	6.18	6.93	1.23	1.46
I-129 (dpm/mL)*	3.70E+02	6.16E+02	6.37E+02	6.34E+02	6.66E+01	5.52E+01

* average of duplicate results

Conclusions

I-129 concentrations above analytical detection limits were observed for six tank farm supernate samples and the overall chemical composition of each sample was determined. The iodine results were consistent with previous results which were above detectable limits. Cesium and technetium concentrations for these samples were reported previously.[4] This data can be added to the iodine inventory estimate currently being developed by Savannah River Remediation (SRR).

References

[1] *Performance Assessment for the H-Area Tank Farm at the Savannah River Site*, SRR-CWDA-2010-00128, Rev. 1, November 2012.

[2] K. D. B. Dixon and M. S. Layton, *Updated Radionuclide and Chemical Inventories in Tanks 9, 10, 11, 13, 14 and 15*, SRR-CWDA-2015-00166, Rev. 1, January 2016.

[3] W. D. King, *Task Technical and Quality Assurance Plan for Reducing SRS Waste Iodine Inventory Uncertainty*, SRNL-RP-2016-00651, Rev. 0, October 2016.

[4] M. D. Fowley, C. J. Bannochie and W. D. King, *Summary of Unreported SRNL Iodine Data*, SRNL-L3100-2016-00221, Rev. 0, December 2016.