WSRC-TR-2002-00540

Reanalysis of Plutonium and Americium-241 in the Tank 19F Closure Grab and Core Samples

R. F. Swingle
Westinghouse Savannah River Company
Aiken, SC 29808

Keywords: Tank Farm, Characterization, Tank closure

Summary

The Tank 19F closure characterization samples (FTF-075, FTF-077 and FTF-118) have been re-analyzed for plutonium isotopes and ^{241}Am. The re-analysis reduced the results for ^{238}Pu by a factor of about two for both the grab samples FTF-075 and FTF-077. The ^{238}Pu results for the core sample FTF-118 remained about the same. The overall average of the three samples was reduced by about 40%. The re-analyzed results for the other isotopes measured remained about the same.

The FTF-075 and FTF-077 grab samples were pulled from Tank 19F in September 2001 as part of the characterization process for closure of Tank 19F.[1,2] The samples were delivered to the Savannah River Technology Center (SRTC) Shielded Cells on September 28, 2001 and placed in the Shielded Cells on October 2, 2001. The samples were opened and both grab samples were found to contain plenty of material to allow completion of the analyses (~70 g in FTF-075 and ~40 g in FTF-077). The samples were dark and resembled marsh muck.

The FTF-118 core sample was pulled from Tank 19F in December 2001 as part of the characterization process for closure of the tank. The sample was delivered to the SRTC Shielded Cells on December 6, 2001 and placed in the Shielded Cells on December 7, 2001. The sample was opened and found to contain plenty of material to allow completion of the analyses (~180 g). The sample resembled a somewhat drier version (more like wet soil) of the previous grab samples FTF-075 and FTF-077.

Chemical analyses of these samples were documented in a previous report.[3] A measurement of the density of the samples was reported in September 2002.[4] This measurement was made after, SRTC personnel suggested that the previously assumed density for the Tank 19F heel (0.234 g/mL) was not appropriate for degraded IE-95 zeolite resin which was believed to be the predecessor for most of the material remaining in Tank 19F.

Once the measured density was incorporated into the final characterization of the heel, it was recognized the concentration values for ^{241}Am and various plutonium isotopes indicated higher heel inventory values than desired. The blank results for the original analysis indicated that there was considerable ^{238}Pu contamination, indicating that at least the concentration for that isotope was probably not as high found in the original analysis. High Level Waste Engineering (HLWE) therefore requested reanalysis of the samples for all of the isotopes whose original analysis gave a higher than desirable heel inventory.[5,6]

Introduction

Tank 19F is scheduled to be closed by March 2004. To close this tank, a characterization of the waste remaining in the tank was required to confirm the inventory of various species for input into groundwater transport models. This characterization has been developed by a combination of process knowledge, visual observation and sample analysis. The characterization samples were obtained by High Level Waste Division (HLWD) personnel and characterized by SRTC personnel.[3,4]

Tank 19F is a 1.3 million gallon capacity Type IV waste tank that was placed in service in 1961. The tank is an 85-feet diameter flat-bottomed cylindrical carbon steel tank with a domed roof. The walls are roughly 35 feet high with the center height of about 45 feet. There are no cooling coils or supports inside the tank.

From July 1980 to July 1981, greater than one million gallons of radioactive waste salt were removed from Tank 19F using mechanical agitation consisting of two 1200-gpm long-shafted jet mixer pumps located in the east and west risers. This method involved adding inhibited water to the tank and stirring the contents with the long-shafted pumps. The resulting salt solution was sent to Tank 18F. Four batch transfers were conducted to remove the salt inventory in Tank 19F. About 98% of the salt and 86% of the radionuclides were removed. Greater than 2,300,000 gallons of water were added to process the four batches.[7]

At the completion of the salt removal campaign, approximately 33,000 gallons of solids remained in an hourglass-shaped formation (running north to south) on the tank bottom. The heel composition was estimated to be 13,000 gallons of spent zeolite resin, 7,000 gallons of metal oxides/hydroxides (standard sludge), and 13,000 gallons of solid salts.[7] The origin of the zeolite in the tank can be traced to a cesium removal column that once operated above the northeast riser to remove cesium from the evaporator overheads. Zeolite was used as filter material for this stripper column. When it needed to be replaced, the old zeolite was dropped into Tank 19F. It was assumed that none of the zeolite was transferred to Tank 18F during the salt removal campaign.

A second waste removal campaign from September 2000 to June 2001 reduced the heel volume from 33,000 gallons to approximately 15,000 gallons.[8,9] In September of 2000 three Flygt mixers were placed into the East, Southwest, and West risers of Tank 19F. The purpose of the mixers was to suspend, mix, and mobilize the waste contents of Tank 19F in preparation for waste transfer into Tank 18F. A 185 gpm centrifugal Bibo transfer pump was installed in the Northeast riser of Tank 19F. A Goulds centrifugal transfer pump was installed in the West riser of Tank 18F to transfer supernate back to Tank 19F. This supernate recycle loop minimized process water addition in order to conserve the limited tank space available. The heel removal strategy consisted of a standard batch transfer and solids suspension operation to remove the Tank 19F solids to Tank 18F. Decanted supernate from Tank 18F was used as the slurry media source. Multiple Flygt mixer orientations and schemes based on testing and experience in Tank 19F were used to provide the maximum removal of the Tank 19F solids.

Discussion

Experimental

Shielded Cells Operations personnel prepared the sample as directed by Waste Processing Technology (WPT) and Analytical Development Section (ADS) personnel in written instructions.

The solid samples, the fresh FTF-118 core samples and the inhibited water-contacted FTF-075 and FTF-077 grab samples, were dried to a constant weight (heated to ~100-115°C and dried until no more weight is being lost due to water evaporation). Portions of the dried solids samples were dissolved in duplicate using a microwave-acid (nitric + hydrofluoric acid) dissolution for a total of two digested samples from each solids sample. The dissolutions were made using ~1 g of solid material in 10 mL of liquid. This is considerably more concentrated than the usual 0.25 g of solid in 250 mL of liquid. This

more concentrated dissolution was made to help in distinguishing between radionuclides actually present in the sample and those introduced by contamination or interference.

The dissolved solid samples were treated with an ion-exchange resin (AMP-1) to remove the Cs-137 to allow removal from the Shielded Cells and submittal to ADS for analyses. In addition to allowing removal of the more concentrated sample from the Cells, this treatment had the potential added benefit of removing the interference of the concentrated Cs-137 from the sample. The samples were removed from the Cells and submitted to ADS for analysis. Two blank dissolutions were also provided. The blanks were also subjected to the Cs-137 removal step.

The analytical techniques that were used for radionuclide analysis include gamma counting for gamma-emitting isotopes (Am-241). Inductively Coupled Plasma – Mass Spectrometry (ICP-MS) was used to determine the concentration of Pu-239 and Pu-240. A chemical separation followed by alpha counting was used to determine the Pu-238 and Pu-241 concentrations.

Results

The results of the reanalysis are given in Table 1. The results for each sample are given as the average and standard deviation of two determinations. For comparison the overall average for the previous determination is also given.[3] In all cases the blanks for the current determinations indicated no significant contamination compared with the actual samples.

The new analyses give about 40% lower ^{238}Pu concentrations, along with a much lower standard deviation. The ^{241}Pu concentrations are also much lower because we now have an actual value for the grab samples (FTF-075 and FTF-077) instead of just a lower limit of detection. The new ^{241}Am concentrations are comparable to those previously measured. The new ^{239}Pu values are somewhat higher than previously measured, and the ^{240}Pu concentrations are somewhat lower due again to the fact that the new measurement gave a real value instead of a lower limit of detection.

Table 1. Results of the Reanalysis of Tank 19 Grab and Core Samples for Pu and ^{241}Am

	FTF-075 (µ Ci/g)		FTF-077 (µ Ci/g)		FTF-118 (µ Ci/g)	
Species	**Average**	**Std. Dev.**	**Average**	**Std. Dev.**	**Average**	**Std. Dev.**

^{238}Pu	0.377	0.010	0.253	0.045	0.285	0.048
^{238}Pu (previous)	0.633	0.502	0.612	0.487	0.289	0.069
^{241}Pu	0.941	0.107	0.810	0.040	0.794	0.034
^{241}Pu (previous)	<6.14	-	<6.71	-	0.902	0.205
^{241}Am	0.179	0.011	0.126	0.007	8.34E-02	4.1E-03
^{241}Am (previous)	0.162	0.027	0.123	0.019	0.138	0.021
^{239}Pu	0.489	0.097	0.400	0.031	0.315	0.012
^{239}Pu (previous)	0.265	0.036	0.243	0.027	0.308	0.079
^{240}Pu	0.169	0.032	0.142	0.011	0.107	0.013
^{240}Pu (previous)	<0.250	-	<0.252	-	7.97E-02	1.64E-02

Quality Assurance

The work documented herein was performed under an approved task technical and quality assurance plan. Shielded Cells Operations personnel, according to written instructions provided by WPT and ADS personnel conducted all sample preparation work. All results were recorded in laboratory notebook WSRC-NB-2002-00191.

Acknowledgments

The author gratefully acknowledges the assistance of D. J. Wheeler, A. L. Williams, C. J. Coleman and D. P. Diprete in the completion of this work.

References

1. N. Elraheb, "Tank 19 Closure Sample Characterization", HLE-TTR-2001-072, Revision 0, August 22, 2001.

2. R. F. Swingle, "Task Technical, Quality Assurance and Characterization Plan for the Tank 19F Tank Closure Samples", WSRC-RP-2000-00799, Revision 1, September 21, 2001.
3. R. F. Swingle, "Characterization of the Tank 19F Closure Grab and Core Samples and the Tank 18F Dip Sample," WSRC-TR-2002-00107, Revision 0, March 6, 2002.
4. R. F. Swingle, "Densities of the Tank 19F Closure Grab and Core Samples," WSRC-TR-2002-00453, Revision 0, September 30, 2002.
5. J. L. Thomas, Measurement of Tank 19 Heel Material Density and Re-Analysis of Am and Pu Concentrations," HLE-TTR-2002-122, Revision 1, October 14, 2002.
6. R. F. Swingle, "Task Technical, Quality Assurance and Characterization Plan for the Re-Analysis of Tank 19F Tank Closure Samples," WSRC-RP-2002-00554, Revision 0, October 24, 2002.
7. C. R. Hayes, "Sampling, Analysis and Characterization Plan to Support HLW Tank 19 Closure", HLW-REG-2000-00033, Rev. 1, August 30, 2001.
8. "Tank 19 Closure Strategy", HLW-WRE-2001-00048, August 16, 2001.
9. J. L. Thomas, "Tank 19 Heel, Supernate, and Possible Unmoved Heel Mounds Volume Calculation," G-CLC-F-00180, Revision 1, February 25, 2002.