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Where is the Plutonium?: Detection and Location of Plutonium-Containing Particles In Tank 18 Waste Using Scanning Electron Microscopy (SEM), Energy Dispersive Spectroscopy (EDS), Wavelength Dispersive Spectroscopy (WDS), and X-ray Diffraction (XRD)

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The F-Area Tank Farm (FTF) Performance Assessment (PA) utilizes waste speciation in the waste release portion of the FTF fate and transport model for movement of chemical and radionuclide into the environment. The waste release modeling associated with the residual plutonium in Tank 18 has been identified as a primary contributor to the Tank 18 dose uncertainty. In order to reduce the uncertainty related to plutonium in Tank 18, a better understanding of the plutonium speciation in the Tank 18 waste (including the oxidation state and stoichiometry) is desired. Savannah River National Laboratory (SRNL) utilized Scanning Electron Microscopy (SEM), Energy Dispersive Spectroscopy (EDS), Wavelength Dispersive Spectroscopy (WDS), and X-ray Diffraction (XRD) to analyze Tank 18 samples to provide information on the speciation of plutonium in the waste material.

Samples from Tank 18 were air dried, ground using a mortar and pestle, and sieved through a 30 mesh (600 µm) screen. The samples for the SEM/EDS/WDS were mounted in epoxy and ground/polished to reveal cross-sectional views of the particles in the sample.

XRD analysis of the Tank 18 samples did not identify any plutonium mineral phases in the samples. This indicates the crystalline mineral phases of plutonium were below the detection limits of the XRD method or that the plutonium phase(s) lacked long-range order and were present as amorphous or microcrystalline solids.

The plutonium identified in the samples was in the form of discrete particles usually <1 µm, but ranging up to several micrometers in diameter. The plutonium particles were spread unevenly within an iron-rich matrix. Due to the small size and low concentration of the plutonium particles, the chemical form of the plutonium remains uncertain. The scan of the small plutonium particles invariably included some of the background iron matrix as a result of the larger interaction volume of the electron beam. Qualitatively, the particles of plutonium found in the SEM analysis do not appear to account for all of the plutonium in the sample based on analysis of the Tank18 samples [1]. A simplified calculation that uses the volume interrogated by the SEM, relevant sample characteristics, and some simplifying assumptions indicated that roughly a million ~1 µm plutonium particles should be visible in an image such as figure 1. The assumptions used in the calculation include: plutonium particles present as homogenously distributed PuO_2 with a diameter of 1 µm and a density of 11.5 g/cm^3, a molecular mass of 276 g/mole, a plutonium concentration of 250 mg/kg in the sample, a bulk sample density of 2.0 g/cm^3, and an SEM sampling depth of 10 µm. This suggests that plutonium is also distributed throughout the solids in low concentration. Note in figure 1 that there do not appear to be any individual plutonium particles, but WDS analysis show some sampled locations of plutonium above the limit of detection (LOD).

References:

[1] L. N. Oji, D. Diprete, and C. J. Coleman, "Characterization of Additional Tank 18F Samples". SRNL-STI-2010-00386, Rev. 0, September 2, 2010.
[2] The authors acknowledge funding from the U.S. Department of Energy.

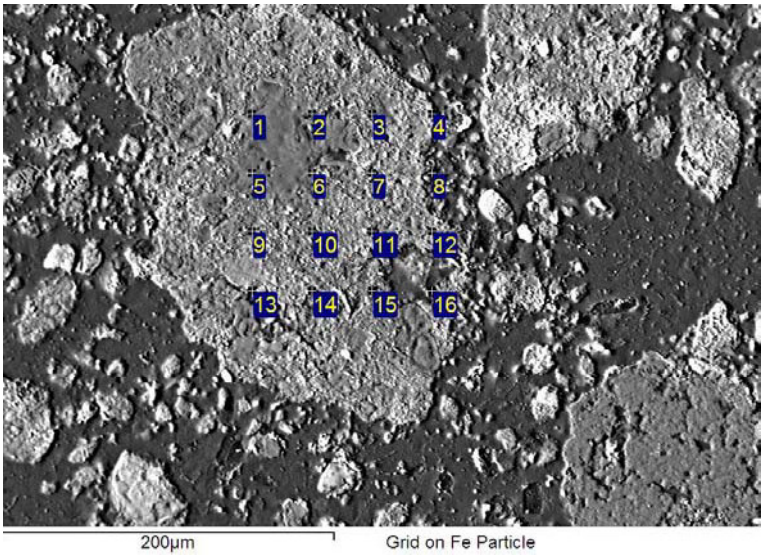

Figure 1. Grid of analysis locations on iron particle.

	EDS Wt %[1,2]									WDS Wt %[3]
Spot	**Na[1]**	**Mg[1]**	**Al[1]**	**Si[1]**	**Ca[1]**	**Mn[1]**	**Fe[1]**	**U[1]**	**O[1,2]**	**Pu[3]**
1	2.1	9.7	13	5.6	6.2	2.3	20	5.2	35	< LOD
2	1.5	6.1	17	5.4	7.0	2.0	19	5.3	36	< LOD
3	5.3	7.8	17	13	0.79	1.5	6.2	8.0	41	0.097
4	7.3	7.0	15	2.2	3.8	1.4	16	15	32	0.19
5	0.0	3.6	21	2.2	1.5	2.2	32	3.6	34	< LOD
6	0.10	7.9	13	2.4	3.3	3.8	32	4.4	32	< LOD
7	5.4	6.9	6.6	1.6	1.2	1.6	13	37	26	< LOD
8	5.7	7.4	15	7.1	2.9	2.9	12	9.8	36	0.073
9	2.6	5.9	12	2.1	9.1	2.2	25	8.4	31	< LOD
10	2.0	13	13	3.3	2.1	2.7	24	5.3	34	0.082
11	1.0	16	22	2.6	1.4	1.1	11	5.8	39	0.11
12	4.5	11	12	2.5	1.7	2.4	24	9.6	32	0.14
13	4.8	6.7	14	6.6	1.6	2.3	19	9.2	35	< LOD
14	4.5	7.6	11	3.2	11	4.8	16	8.6	32	0.13
15	2.3	11	16	5.0	3.7	1.9	19	4.8	36	< LOD
16	4.6	4.2	26	2.2	0.91	1.4	17	6.7	37	< LOD
[1]EDS numerical results are for trend analysis only and are normalized to 100%. EDS results are semi-quantitative estimates based on standardless analysis and theoretical intensity corrections from Oxford Instruments INCA 4.15 EDS software.										
[2]Oxygen calculated by stoichiometry, assuming Na_2O, MgO, Al_2O_3, SiO_2, CaO, MnO, Fe_2O_3, UO_2. Pu is not included in the oxygen calculation.										
[3]Pu Limit of Detection (LOD) is ~0.04 wt% based on 3-sigma counting statistics. Pu estimated uncertainty +/- 30% for values < 0.1 wt% based on 2-sigma counting statistics.										

Table 1. Elemental energy dispersive spectroscopy (EDS) and wavelength dispersive spectroscopy (WDS) weight % for selected locations from Figure 1.

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