SRNL
SAVANNAH RIVER NATIONAL LABORATORY

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Technical Review: Kent H. Rosenberger Management Concurrence: B. T. Butcher

Estimation of the Potential Contamination on Corrosion Products in Tank 18

Introduction and Summary

SRNL has been asked by High Level Waste to estimate the amount of radioactive contamination that might be found on the interior walls of Tank 18. Data was requested from HLW on the composition of the liquid in Tank 18 when it was in service. B. J. Weirsma, of SRNL-MTS provided an estimate of the mass of corrosion products in the tank.

Methodology

When a solution is in contact with a solid phase, the constituents partition between the solid and liquid. The factor used to quantify this is called the partition coefficient, or K_d. It is defined as the concentration in the solid phase divided by the concentration in the liquid phase and has the units volume/mass:

$$K_d = (Ci/Kg)/(Ci/L) = L/Kg \qquad \text{Eq. 1}$$

In the High Level Waste Tanks system, liquid tank contents are in contact with corrosion products in the tank walls. The question to be answered is how much radioactive contamination could be held on the corrosion products. K_d values for many chemical species have been measured for hydrous iron oxides (FeOOH, goethite, rust). Given information on the liquid concentrations, K_ds and the mass of corrosion products, the K_d definition equation can be used to answer this question.

$$Ci_{solid} = K_d * (Ci/L)_{liquid} * Kg_{solid} \qquad \text{Eq. 2}$$

Analytical data from a dip sample taken from Tank 18 in 1975 (Attachment A) was used to represent the solution that was in contact with the tank walls when the tank was in service. These data are given in Table 1.

The partition coefficients for the elements in question on iron oxide (goethite) for an alkaline environment are given in Table 2.

The Materials Technology Section of SRNL gave an estimate of 724 lb (329 Kg) of rust on the walls of the tank (Wiersma 2003).

Table 1. 1975 Sample Analytical Results for Tank 18

Radionuclide	Concentration, Ci/L[a]
^{3}H	1.85E-05
^{14}C	1.75E-06
^{60}Co	1.99E-08
^{63}Ni	2.03E-06
^{79}Se	9.85E-08[b]
^{90}Sr	1.68E-06
^{94}Nb	1.74E-12
^{99}Tc	4.49E-04
^{129}I	6.29E-08
^{134}Cs	1.45E-07
^{137}Cs	2.01E-01
^{233}U	1.84E-06
^{234}U	1.19E-06
^{235}U	1.06E-09
^{236}U	1.23E-08
^{238}U	2.69E-08
^{237}Np	1.34E-07
^{238}Pu	3.84E-07
^{239}Pu	1.18E-05
^{240}Pu	4.32E-05
^{241}Pu	4.93E-07
^{242}Pu	7.24E-07
^{241}Am	1.41E-06

[a] Derived from data provided by HLW (Attachment A), decay corrected to 2005

[b] Derived from P. Hill E-mail - Attachment A

Table 2. Partition Coefficients for Goethite (FeOOH) in an Alkaline Environment

Element	K_d, L/Kg
H	0 (Table E.4-1, WSRC, 2000)
C	0 (Table E.4-1, WSRC, 2000)
Co	10 (Bradbury and Sarott, 1995)
Ni	10 (Bradbury and Sarott, 1995)
Se	2 (Cook, 2005)
Sr	3 (Table E.4-1, WSRC, 2000)
Nb	50 (Bradbury and Sarott, 1995)
Tc	0 (Table E.4-1, WSRC, 2000)
I	0 (Table E.4-1, WSRC, 2000)
Cs	5 (Ohnuki, 1991)
U	6000 (Table E.4-1, WSRC, 2000)
Np	750 (Table E.4-1, WSRC, 2000)
Pu	2000 (Table E.4-1, WSRC, 2000)
Am	3700 (Table E.4-1, WSRC, 2000)

Combining the results in Tables 1 and 2 and the mass of rust from MTS, according to Equation 2 gives an estimate of the radioactive contamination that might be on the walls of Tank 18. These results are shown in Table 3.

It should be noted that the K_d for H, C, Tc, and I on goethite under alkaline conditions is 0 L/Kg, meaning that these radionuclides would not be expected to be associated with corrosion products in a high level waste tank operated under high pH conditions.

Table 3. Estimate of Contamination on Tanks 18 Walls

Radionuclide	Activity, Ci
^{3}H	0
^{14}C	0
^{60}Co	6.6E-05
^{63}Ni	6.7E-03
^{79}Se	6.5E-05
^{90}Sr	1.7E-03
^{94}Nb	2.9E-08
^{99}Tc	0
^{129}I	0
^{134}Cs	2.4E-04
^{137}Cs	3.3E+02
^{233}U	3.6E+00
^{234}U	2.3E+00
^{235}U	2.1E-03
^{236}U	2.4E-02
^{238}U	5.3E-02
^{237}Np	3.3E-02
^{238}Pu	2.5E-01
^{239}Pu	7.7E+00
^{240}Pu	2.8E+01
^{241}Pu	3.2E-01
^{242}Pu	4.8E-01
^{241}Am	1.7E+00

Conclusions

The radionuclide inventories shown in Table 3 are compared to the estimates of the residual heel (sludge plus supernate) in Tank 18 in Table 4. Several of the actinide radionuclides (^{233}U, ^{234}U, ^{236}U, and ^{242}Pu) have larger calculated inventories on rust than are estimated to be in the heel. The magnitude of the inventories of these four radionuclides on the rust is quite low, and previous modeling work indicates that these inventories would produce no measurable effect at the point of assessment (USDOE 1996).

Table 4. Comparison of Estimated Wall Residual and Bottom Residual in Tank 18

Radionuclide	Wall Residual, Ci	Residual Heel, Ci[a]
^{60}Co	6.6E-05	4.4E+00
^{63}Ni	6.7E-03	8.0E+01
^{79}Se	6.5E-05	5.0E-02
^{90}Sr	1.7E-03	1.4E+03
^{94}Nb	2.9E-08	4.0E-05
^{134}Cs	2.4E-04	3.1E-03
^{137}Cs	3.3E+02	1.2E+04
^{233}U	3.6E+00	8.1E-01
^{234}U	2.3E+00	2.3E-01
^{235}U	2.1E-03	7.2E-03
^{236}U	2.4E-02	7.5E-03
^{238}U	5.3E-02	1.8E-01
^{237}Np	3.3E-02	8.6E-02
^{238}Pu	2.5E-01	7.0E+01
^{239}Pu	7.7E+00	1.3E+02
^{240}Pu	2.8E+01	3.0E+01
^{241}Pu	3.2E-01	2.5E+02
^{242}Pu	4.8E-01	8.0E-02
^{241}Am	1.7E+00	7.2E+01

[a] From Tran, 2005

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ATTACHMENT A

Tank 18 Supernate Estimates

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Tank 18 Supernate Estimate from HLW – 4/18/2--5

	Current Projections			Projected for old sample, Ci			Best Value for Old Sample, Ci	Best Value for Old Sample, Ci/gal
	WCS	recent sample	combine	WCS	recent sample	combine		
H-3	2.31E+00	6.16E-02	6.16E-02	2.31E+00			2.31E+00	3.79E-04
C-14	4.06E-02		4.06E-02	4.06E-02			4.06E-02	6.66E-06
Co-60	2.38E-02		2.38E-02	2.38E-02			2.38E-02	3.90E-06
Ni-59								
Ni-63	5.77E-02		5.77E-02	5.77E-02			5.77E-02	9.46E-06
Se-79								
Sr-90	2.86E-02		2.86E-02	2.40E+00			7.99E-02	1.31E-05
Y-90	2.86E-02		2.86E-02				7.99E-02	1.31E-05
Nb-94	4.01E-08		4.01E-08	4.01E-08			4.01E-08	6.58E-12
Tc-99	1.23E-01	9.43E-03	9.43E-03	1.03E+01			1.03E+01	1.70E-03
Ru-106							1.35E+02	2.21E-02
Rh-106								
Sb-125								
Sn-126								
I-129	2.08E-07		2.08E-07	1.45E-03			1.45E-03	2.38E-07
Cs-134							7.89E+01	1.29E-02
Cs-135								
Cs-137	1.17E+02	1.32E+00	1.32E+00				9.24E+03	1.52E+00
Ba-137m	1.10E+02	1.25E+00	1.25E+00				8.74E+03	1.43E+00
Ce-144							8.72E+01	1.43E-02
Pr-144								
Pm-147								
Eu-154								
Th-232								
U-232	4.00E-07		4.00E-07	1.98E-07		2.03E-07		
U-233	**1.74E-02**	1.44E-02	1.44E-02	8.65E-03	4.23E-02	7.30E-03	4.23E-02	6.95E-06
U-234		9.30E-03	9.30E-03		2.73E-02	4.71E-03	2.73E-02	4.49E-06
U-235	3.03E-06	8.30E-06	8.30E-06	1.50E-06	2.44E-05	4.21E-06	2.44E-05	4.01E-09
U-236		9.62E-05	9.62E-05		2.83E-04	4.88E-05	2.83E-04	4.64E-08
U-238	1.71E-04	2.10E-04	2.10E-04	8.50E-05	6.19E-04	1.07E-04	6.19E-04	1.02E-07
Np-237	**1.75E-03**	1.05E-03	1.05E-03	8.69E-04	3.09E-03	5.32E-04	3.09E-03	5.06E-07
Pu-238	7.26E-01	3.81E-03	3.81E-03	3.60E-01	1.12E-02	1.93E-03	1.12E-02	1.84E-06
Pu-239	1.04E-01	9.25E-02	9.25E-02	5.15E-02	2.72E-01	4.69E-02	2.72E-01	4.46E-05
Pu-240	2.61E-02	3.39E-01	3.39E-01	1.30E-02	9.97E-01	1.72E-01	9.97E-01	1.64E-04
Pu-241	7.43E-01	1.66E-02	1.66E-02	3.69E-01	4.87E-02	8.39E-03	4.87E-02	7.99E-06
Pu-242	3.55E-05	5.67E-03	5.67E-03	1.76E-05	1.67E-02	2.88E-03	1.67E-02	2.74E-06
Ingrown Am-241	8.11E-02		8.11E-02	4.02E-02		4.11E-02		
Am-241	**8.20E-02**	1.16E-02	1.16E-02	4.07E-02	3.40E-02	5.87E-01	3.40E-02	5.59E-06
Am-242m	**8.03E-01**		8.03E-01	3.99E-01		4.07E-01		
Cm-244	3.43E-01		3.43E-01	1.70E-01		1.74E-01		
Cm-245	3.97E-12		3.97E-12	1.97E-12		2.01E-12		

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cc
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Subject
Tank 18 Se-79 in Rust

Based on process history information, the bottom 160" of Tank 18 was exposed to material with an average Se-79 concentration of 3.73 E-07 Ci/gal

Distribution

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W. E. Stevens, 773-A
B. T. Butcher, 773-43A
L. R. Bickford, 730-A
C. F. Jenkins, 730-A
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H. Q. Tran, 766-H
K. H. Rosenberger, 766-H
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