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**Response to the NRC's Request for Additional Information Comment #15**

The Savannah River Site draft 3116 Determination (DOE-WD-2005-002) was provided to the Nuclear Regulatory Commission on September 30, 2005 for review. The NRC issued a Request for Additional Information on March 31, 2006. The attachment to this memorandum contains the response to the Request for Additional Information comment 15. The NRC's original text has also been reproduced for your convenience.

**NRC COMMENT #15** INVENTORY AND SAMPLING

Inventory estimates are developed through a combination of sampling, process knowledge, and special calculation. Justification that process knowledge and special calculation can provide reliable predictions is not provided.

**BASIS**

Tables 2-3 and 2-4 of the draft waste determination provide the estimated residual inventory for Tanks 18 and 19. There are values in the tables that are not intuitive, and the Waste Characterization System (WCS) is not amenable to independent verification without recourse to the developers. For example:

- The sampled values for uranium isotopes range from a factor of four to 30 less in Tank 19 compared to Tank 18; however, the WCS generated value for U-232 is nearly identical.
- The Cm-244 value in Tank 19 generated with the WCS is roughly five orders of magnitude less than the value for Tank 18, even though the other Cm isotopes are comparable.
- The Pu-244 value (from special calculation) in Tank 19 is roughly four times higher than the value for Tank 18; however, all the sampled isotopes of Pu are less in Tank 19 than Tank 18.
- The Characterization of Tank 19 Residual Waste [15] indicates that, prior to sampling, the concentration of Cs-137 in Tank 19 solids was underpredicted by factor of approximately 200. The underprediction is discussed and attributed to partitioning of Cs-137 onto zeolite. However, it is not clear that Cs-137 is the only radionuclide that is likely to partition onto the zeolite, and the possible underprediction of the concentration of unsampled radionuclides in Tank 19 solids due to partitioning onto zeolite is not discussed.
- The Characterization of Tank 19 Residual Waste [15] indicates that, prior to sampling the concentration of Pu-242 in the Tank 19 solids was underpredicted by approximately a factor of 10. No reason for the underprediction is discussed, and no assessment of the possibility that unsampled radionuclides could be underpredicted by a similar factor due to similar processes is provided.

In some cases, estimators such as fission yields are used to estimate the concentrations of radionuclides that are not measured. However, it is not clear why this would be a reliable estimator if tank inventories are accumulated over many years as a result of different operations, and the sampled radionuclide and estimated radionuclide have different chemical behavior in the tank environment.

Table 3-5 of the Performance Objectives Demonstration Document (PODD) provides detailed estimates on a tank by tank basis. The following observations are made:

- Tanks 17 and 19 are estimated (based on sampling) to contain the two highest Tc-99 concentrations in the tank farm. It is unlikely that the Tc-99 inventory in all of the F Tank Farm tanks that have not been sampled is lower than these sampled values, unless there is a reason to expect that Tanks 17 and 19 contain the waste with the highest Tc-99 concentrations in the tank farm. Thus, it seems that the Tc-99 inventories in the other tanks in the tank farm have been underestimated.

- The sampled Tank 18 value for Np-237 is by far the highest value of all tanks. DOE acknowledges that part of the reason for the high Np-237 in Tank 18 is the introduction of laboratory waste that was not tracked with the WCS [18]. The result demonstrates that untracked waste can have a significant impact on estimates generated with the WCS.

- The total inventory of Np-237 in Tanks 17-20 is roughly three times higher than the predicted inventory in Tanks 1-8. However, the concentration of Np-237 in Tanks 17-20 is roughly 30 times lower than the predicted concentration in Tanks 1-8. This suggests that either DOE has assumed roughly 100 times better waste removal for Tanks 1-8 than for Tanks 17-20 when generating the inventory and the overall risk from the F Tank Farm or the concentrations are inaccurate.

- There is zero estimated tritium in Tanks 1-8 and Tanks 25-28, 33-34, and 44-47, although the sampled tank group (Tanks 17-20) has measured tritium.

- The concentration of I-129 is lowest for the tank group of 17-20, as estimated with the WCS, compared to the other tank groups. It is not clear why I-129 concentrations are so much lower for Tanks 17-20 than the other tanks.

These items are identified because they cause concern that the unsampled radionuclides in Tanks 18 and 19 may have a high degree of uncertainty that is not accounted for and that the current inventory of the F Tank Farm presented in the PODD may not be sufficiently accurate for decision making.

**PATH FORWARD**

Describe any studies that have been done to assess the reliability of predictions of inventory based on WCS data. Provide an

explanation of the differences between the WCS generated values and the sampled values, and assess the possibility that unsampled radionuclides could be underpredicted, specifically addressing processes believed to lead to the underprediction of some of the sampled radionuclides. Provide uncertainty estimates for the inventories based on process knowledge and special calculations presented in Tables 3-2 and 3-4 of the PODD. Uncertainties in radionuclide inventories, especially that of Cm-244, should be considered in the comparison of waste concentrations to Class C concentration limits.

**SRS RESPONSE**

SRS has not previously performed studies to compare WCS predictions to tank heel inventories derived from sample results. In response to the NRC's comment, this response examines the Tank 19 and 18 heel inventory predictions made to support tank closure in comparison to the sample-derived inventories. Table 15-1 contains the information examined. Note that the sample-derived inventories in Table 15-1 are based on the average sample results instead of the 95% Upper Confidence Level sample results used in the PODD (CBU-PIT-2005-00106). The third column of Table 15-1 identifies the starting point for each prediction. The starting point for these predictions is either the fission-yield computer code or accountability records. Since none of the unsampled isotopes is characterized by accountability records, the uncertainty of predictions based on accountability records is not germane to this examination. In addition to the information contained in the Tank 19 and Tank 18 residual material characterizations, a retrospective look at the gamma pulse height analysis data from the Tank 19 and Tank 18 material analyses has provided more information for comparison. This data is shown in Tables 15-2a and 15.2b. It can be seen that the only radionuclides present above the detection limit are Co-60 in both tanks and Eu-154 in Tank 18. Additionally, while no definite conclusions can be drawn regarding the accuracy of predicted inventories for radionuclides with sample values less than the minimum lower limit of detection (LLD), for at least 8 of the radionuclides it is clear that the predicted value is higher than the actual value. To further aid in the examination, predictions based on fission-yield calculations are shown separately in Table 15-3. A closer inspection of the fission-yield based predictions in Table 15-3 identifies the following:

1. As previously identified in WSRC-TR-2002-00052 (page 15), Cs-137 and Ba-137m are severely under-predicted due to the effects of Cs-137 accumulation on the zeolite resin.
2. All other fission products with above-detection-limit sample-

based inventories are over-predicted.

The NRC requests that SRS "provide an explanation of the differences between the WCS generated values and the sampled values". There are three main factors that contribute to the WCS-based predictions being higher than the sample derived predictions. First, for conservatism, each reactor assembly is assumed to have received the maximum exposure possible and, therefore, the actual fission products contained in an assembly are less than those entered into WCS. Second, the WCS predictions are obtained by multiplying the WCS radionuclide concentration (Ci/kg) by the residual mass calculated by Thomas (WSRC-TR-2002-00052). The mass of the non-radioactive constituents in sludge is under-estimated in WCS. Since iron and manganese are credited neutron poisons, for criticality control purposes their inventories in the sludge are under-estimated (for conservatism). As a result of the under-estimated total mass, the WCS radionuclide concentrations in sludge are over-estimated on a Ci/kg basis. When these higher than actual concentrations are multiplied by the actual mass, the radionuclide content is over-estimated. Last, some of the residual material originated as cladding waste or other low radionuclide bearing wastes that, for the purposes of conservatism, are characterized as fission product bearing PUREX Low Heat Waste.

The NRC also requests that SRS "assess the possibility that unsampled radionuclides could be under-predicted". Based on these observations, it is unlikely that radionuclide inventories based on fission-yield estimates are under-predicted unless the radionuclide has been artificially accumulated by zeolite resin capture. Based on research (discussed below), the only radionuclides that could be captured in significant quantities by the zeolite used at SRS are Sr and Eu. It can be seen from the information in Tables 15-1 and 15-2 that neither Sr nor Eu is found in the residual samples in excess of what is predicted. In addition, Cs, Np, Pu, and Am displayed a moderate affinity for zeolite. Since Np, Pu, and Am inventories are derived from sample analyses, no adjustment is required to their reported inventories. As mentioned above, Cs-137 is found in excess of predicted values and, taking this into consideration, it is likely that the Cs-134 and Cs-135 heel inventories predicted by WCS are low by the same magnitude that the Cs-137 inventory is under-predicted. This would mean that the Tank 19 heel inventories for Cs-134 and Cs-135 could be a factor of 183 times higher than predicted and for Tank 18 the inventories could be a factor of 40 times higher than predicted. Since the half-life of Cs-134 is 2.065 years, approximately 50 half-lives will occur during the 100 years of institutional control, rendering the impacts of any Cs-134 inconsequential. The maximum contribution to the all-pathways

dose from Cs-135 is only 3.6E-05 mrem/year for F-Tank Farm from the 100 meter well in the Water Table Aquifer; therefore, an increase by even a factor of 200 would also be inconsequential.

**Discussion of Zeolite**

The zeolite used for treatment of SRS waste evaporator overheads was 96% Linde AW-500 (later known as Ionsiv IE-95), which is a mixture of chabazite, erionite, and clay, and 4% Decalso (WSRC-TR-2002-00288). In tank waste, the IE-95 has been observed to degrade to cancrinite/sodalite minerals, including natrodavyne[1], nitrated sodalite[2], hydroxy-cancrinite[3], and cubic nitrated sodalite[4]. These condensed aluminosilicates are closely related to each other, with minor differences in the unit cell size. The open cages of the original zeolites are large hexagonal/rhombohedral shapes that collapse to much smaller cubic and hexagonal unit cells during decomposition. This condensation also corresponds to exclusion of waters of hydration from 12 to 72 in the original zeolite to zero to four in the condensed aluminosilicates. It has also been shown that very high CsOH solution will form the Cs-containing zeolites Cs-substituted chabazite and pollucite[5]. In highly alkaline solutions, the Decalso degrades to a gel.

The original primary function of the AW-500 and Decalso was to remove Cs-137 from a low activity stream, and these materials have a high affinity for cesium. When the zeolite was loaded with Cs-137, it was dumped to the tank. The zeolite appears to have absorbed more Cs-137, and reacted with the highly alkaline salt solution (CBU-PIT-2005-00099). It is plausible that during formation of the condensed aluminosilicates, the cesium was incorporated into the structure and is not easily exchangeable. After condensation, to sorb a cation from solution, the cation would have to substantially dehydrate to enter the unit cell of the aluminosilicate, which is less thermodynamically favorable than the more open cage of the original zeolite.

Simulant studies of radionuclide surrogates have been performed using a synthesized nitrated cancrinite (PIT-MISC-0154). Cesium was observed to partition to the condensed aluminosilicate, but affinity appears to be a function of the initial load of cesium present when the material was initially formed. High initial Cs loading during formation suppressed subsequent ion exchange activity.

[1] $(Na_6[Al_6Si_6O_{24}](Na_2CO_3))$
[2] $(Na_8[Al_6Si_6O_{24}](NO_3)_2)\cdot 4H_2O)$
[3] $(Na_6[Al_6Si_6O_{24}](2NaOH)\cdot 4H_2O)$
[4] $(Na_6[Al_6Si_6O_{24}](2NaNO_3))$
[5] $(CsAlSi_2O_6)$

Perrhenate (surrogate for pertechnetate), selenate, and iodide were not significantly removed from solution by nitrated cancrinite. Additional studies on desorption of cesium from sodalite and cancrinite formed in the presence of simulant containing cesium have been reported (PIT-MISC-0155). Less than 20% of the cesium was leachable from sodalite and less than 55% from cancrinite, although the displacing ion (e.g. Na, K) impacted the leachability. The ion with lowest dehydration energy was most effective at displacing cesium.

Samples obtained from the top of the northeast mound in Tank 19F prior to final waste removal were analyzed by X-Ray Diffraction (XRD) (SRT-LWP-97-111) and found to contain sludge and sodium aluminum nitrate silicate hydrate[6]. None of the original zeolite species, chabazite and erionite, were found. The original zeolites had converted to this nitrated sodalite/cancrinite structure. Similarly, the grab sample from the mound under the northeast riser during final waste removal activities was found to contain a mixture of the same hydrated nitrated sodalite identified earlier[7], and another nitrate exchanged sodalite[8] (WSRC-RP-2001-00410). These results from an actual tank sample are consistent with observations by Jantzen (WSRC-TR-2002-00288) on the degradation of the original zeolites.

Other radionuclides are also known to be sorbed by AW-500 and IE-95. A simulant of a West Valley waste stream was tested using batch equilibrium tests and shown to have the following relative distribution coefficients (PNL-4969). Results were reported as averages of AW-500 and IE-95, since they are the same material.

| | | Rd (mL/g) |
|---|---|---|
| | pH | AW-500 & IE-95 (average) |
| Cs | 10 | 39 |
| | 13 | 34 |
| Sr | 10 | 386 |
| | 13 | 1408 |
| Pu | 10 | 19 |
| Tc | 10 | <1 |
| | 13 | <1 |
| Np | 10 | 2 |

[6] $(Na_8(Al_6Si_6O_{24})(NO_3)_2 \cdot 4H_2O)$
[7] $(Na_8(Al_6Si_6O_{24})(NO_3)_2 \cdot 4H_2O)$
[8] $(Na_8(Al_6Si_6O_{24})(NO_3)_2)$

| | | |
|---|---|---|
| | 13 | 28 |
| I | 10 | 1 |
| | 13 | ~2 |
| Eu | 10 | 790 |
| | 13 | 9300 |
| Ru | 10 | <1 |
| | 13 | 1 |
| Am | 10 | 24 |

Where Rd = $(C_{solid}/C_{liquid})$x(mL liquid/g solid*F);

F is a correction to dry weight of the zeolite.

The highest affinity in this matrix is for Eu, followed by Sr. The relatively moderate affinity for Cs in this matrix is attributable to very high sodium content (7 M) of this simulant, which competes with cesium for cation sites on the zeolite. The sorption of anions Tc (pertechnetate) and I was very low, as expected for anions on zeolites.

Samples of solids from the top of the mound in Tank 19F prior to removal of the waste mound were analyzed (WSRC-RP-97-0074). Although this sample was from the surface of the waste prior to final waste removal, its characteristics were similar to samples during and after waste removal, and it was estimated to contain 30% zeolite based on silicon analysis. The solids were shown to contain Cs-137, Sr-90, Tc-99, and Pu-238/239, but below detection limit concentrations of Sn-126, Sm-151, Eu-154/155, Co-60, Zr-95, U-233/234/236, Am-241, Pu-241/242, and Cm-244, and only trace amounts of Np-237. Later samples collected from under the NE riser during final waste removal were lower for Cs-137, Sr-90, Tc-99, and Pu-238, but uranium, neptunium, and other plutonium species were comparable (WSRC-RP-2001-00410). The 1997 sample results also agreed well with the residual material sample averages (CBU-PIT-2005-00068) used to characterize the Tank 19 residual heel. These data indicate that the solids are comparable, and that the only analyzed radionuclide that is unpredictably high is Cs-137 (Table 15-4).

**Conclusions Pertaining to Zeolite**

In high ionic strength alkaline solutions, the original zeolites are good absorbers for strontium and europium, moderate absorbers for cesium, plutonium, neptunium, and americium, and poor for technetium, iodine, and ruthenium. Since iodine and technetium are

present as anions in waste, there is no expectation that they would be exchangeable for the sodium in the original zeolite.

The original zeolites degrade in highly alkaline solution to condensed aluminosilicates with low unit cell dimensions. If the original zeolites were not good absorbers for particular radionuclides, it is not expected that the condensed aluminosilicates would contain appreciable amounts because (a) they were not present in high concentrations during condensation/formation and (b) the small unit cell dimensions will limit ion transport afterwards. Experimental data on simulants confirms that iodine, perrhenate, and selenate are not appreciably exchanged, and cesium exchange is hindered.

The Tank 19F samples contain the condensed aluminosilicates, along with moderate amounts of cesium, and plutonium. Only minimal amounts of other radionuclides that are known to have an affinity for the original zeolites (europium, neptunium) are observed. Several other radionuclides are also not present in the condensed aluminosilicates in appreciable quantities (tin, samarium, cobalt, zirconium). This indicates that the condensed aluminosilicates are not good absorbers for these species, or never encountered appreciable quantities of them.

While these conclusions do not unequivocally prove that other un-analyzed radionuclides are not present in the tank solids, they do indicate that there is no reason to suspect that they were present during zeolite decomposition or are selectively sorbed by the condensed aluminosilicates.

## Uncertainty Estimates for Process Knowledge and Special Calculations

In order to develop uncertainty estimates for inventories based on process knowledge and special calculations presented in Tables 3-2 and 3-4 of the PODD (CBU-PIT-2005-00106, pages 97 and 106), it is necessary to understand the basis for each of the estimates.

### Liquid Predictions

The only three radionuclides for which predicted liquid inventories contributes significantly to the total residual inventories in each tank are I-129, Am-242m, and C-14.

The Tank 19 and Tank 18 residual liquid I-129 inventories are based on a recommended I-129 to Cs-137 ratio of 1.57E-7 curies of I-129 per curie of Cs-137 (CBU-PIT-2005-00050). The highest I-129 to Cs-137 ratio reported in the study is 1.63E-07 curies of I-129 per curie of Cs-137 which is less than 4% higher than the ratio used. Since the Tank 18 residual liquid inventory contributes only 3% of

the total I-129 remaining in that tank, increasing the residual liquid inventory of I-129 by 4% would have a negligible impact. The Tank 19 predicted residual liquid inventory of I-129 contributes 89% of the total I-129 remaining in that tank. It is worth noting that the predicted Tank 19 residual liquid I-129 inventory is obtained by multiplying the recommended ratio by the predicted Cs-137 inventory, which is 16% higher than the sample-derived Cs-137 inventory. Although a more conservative ratio could be used to increase the Tank 19 residual I-129 inventory by less than 4%, the contribution to the Tank 19 all-pathways dose from I-129 is only 0.21% of the total dose, so any increase would be inconsequential.

The Tank 19 and Tank 18 residual liquid Am-242m inventories are based on the average concentration (3.48E+04 pCi/ml) of six recent supernate samples (X-ESR-G-00004, page 11). The sample results are reproduced in Table 15-5. As can be seen in Table 15-5, all of the sample results were less-than-detection values. It can also be seen that a single abnormally high detection limit on the Tank 49 sample result skews the average higher than the next highest detection limit. A more realistic but still conservative algorithm would be to exclude the Tank 49 result and use a constant value equal to the average of the 5 remaining results, which is 5.4E+03 pCi/ml. If this more appropriate value was used, the resulting residual inventories of Am-242m in Tank 19 and Tank 18 would be more than a factor of 6 less than the current predicted inventory.

Carbon-14 is formed when nitrogen impurities in aluminum used for cladding and alloyed with uranium for fuel undergo activation. Aluminum in target assemblies (as cladding only) is discarded as PUREX Low Heat Waste (LHW), whereas aluminum in the fuel assemblies is discarded as H-Modified High Heat Waste (HM HHW). The WCS algorithm for C-14 in sludge is based on the average C-14 concentration in these two streams, determined based on analytical data from sludge samples (DPST-83-2001). The analytical data in the report has a relative standard deviation of 27%. In theory, the C-14 inventories in the Tank 19 and Tank 18 solids heels could be up to 54% higher. The majority (over 80%) of the combined Tank 19 and Tank 18 C-14 heel inventory is due to C-14 in the liquid phase. The C-14 residual liquid prediction is derived using the average value of eight recent supernate sample analyses (X-ESR-G-00004, p. 11). The analytical data is reproduced below in Table 15-6. For the purposes of developing the average, the less-than-detection values are assumed to be the actual value. The average of these eight samples is 1.76E+03 pCi/ml, and this value is used to calculate the liquid inventories in Tank 19 and Tank 18. It is worth noting that the value used is actually higher than either of the detected values and that the average of the two detected values is

989 pCi/ml, which is only 56% of the value used. In addition, since the Tank 18 residual liquid was inhibited water instead of supernate, the actual C-14 liquid concentration is expected to be orders of magnitude lower. In the most pessimistic case, the highest less-than-detection value of 7.11E+03 pCi/ml could be used, which would increase the predicted liquid inventory by a factor of 4. Although the estimated inventory is based on average concentrations for the solids and liquid phases, the contribution to the all-pathways dose from C-14 is only 0.10% for Tank 18, <0.01% for Tank 19, and <0.01% for F-Tank Farm; therefore, an under-prediction of the C-14 solids inventory by 54% and the liquid inventory by a factor of four would be inconsequential.

**Solids Predictions**

Although Co-60 is an activation product, the amount of Co-60 that is generated from activation of nickel impurities was included in the computer code that calculated fission yields for SRS reactors (WSRC-TR-94-0562, p. 8). The gamma pulse height analysis data that is displayed in Tables 15-2a and 15-2b contains detectable sample concentrations for Tanks 18 and 19. In both cases the predicted inventories exceed the sample-based inventory.

Nickel-59 and Ni-63 are also activation products whose concentration in SRS waste is calculated using the same methodology as Co-60 (DPST-82-236). A review of the data in Table III (DPST-82-236, p. 8) identifies that, for the assemblies processed in F-Area (Mk31A and Mk31B), the Ni-59 and Ni-63 concentrations was only 60% of the average concentrations that are used by WCS. Therefore, it is expected that both the Ni-59 and Ni-63 inventories are over-predicted.

The inventories of Nb-94, Ru-106, Rh-106, Sn-126, Sb-125, I-129, Ce-144, Pr-144, Pm-147, Eu-154, U-232, Am-242m, and Cm-245 are based on fission yield computer code calculations, so as with the fission-yield predictions discussed above, it is unlikely that the inventories of any of these radionuclides is under-predicted.

Because the inventories of Te-125m, Sb-126m, Sb-126 are calculated based on decay from the fission-yield predicted inventories of Sb-125 and Sn-126, it is unlikely that the inventory of any of these radionuclides is under-predicted. The methodology for calculating Te-125m, Sb-126m, and Sb-126 is described in CBU-PIT-2005-00034.

The Tank 18 predicted Cm-244 inventory is based on a combination of fission-yield computer code calculations and sampling. Curium-244 production is calculated by the fission-yield computer code, so for the majority of transfers into Tank 18, Cm-244 was likely over-

predicted. These transfers total less than 0.1 curies of Cm-244 transferred to Tank 18. In January 1973, a portion of the Site's Cm-244 reserve was declared excess. The canyon vessel containing the excess was sampled, and then the material was transferred to Tank 17. Based on the sample results, 11,811 curies of Cm-244 were transferred to Tank 17. All but 57 curies of this Cm-244 was subsequently transferred to Tank 18 during Tank 17 waste removal efforts conducted between 1979 and 1997. This Cm-244 addition to Tank 17 and the subsequent transfers to Tank 18 are tracked in WCS. Additionally, since this discard was not made as part of normal canyon transfers, it is tracked in WCS as a discrete addition of 11,811 curies that do not decay. Considering that Cm-244 has an 18.1 year half-life and over 33 years has elapsed since the discard, the Cm-244 inventory is over-predicted by approximately a factor of four. Since the Tank 19 Cm-244 inventory is based on fission yield only, it is most likely over-estimated.

The predicted inventories of Sm-151, Eu-152, Eu-155, Am-243, Cm-242, Cm-243, Cm-247, Cm-248, Bk-249, and Cf-249 are based on a combination fission-yield computer code calculations and sample results. For these radionuclides, the fission-yield activity ratio to Sr-90 for 30-year-old waste is determined. These ratios are reproduced below in Table 15-7. The ratios are multiplied by the Sr-90 inventory derived using the 95% Upper Confidence Level (UCL) sample result. This methodology is described in CBU-PIT-2005-00068.

The predicted inventory of Pu-244 is calculated based on the sample derived inventory of Pu-242. The mass of Pu-244 in a tank is very conservatively assumed to be equal to the mass of Pu-242 in the tank as determined by sample results (derivation of this methodology can be found in CBU-PIT-2005-00039). Therefore, the predicted inventory of Pu-244 is certain to be over-predicted. An example of this calculation can be found in CBU-PIT-2005-00068.

The predicted inventories of Th-232 and its daughter Ra-228 are based on accountability records. Since THOREX processing occurred only in H Canyon, the inventories of these radionuclides in Tanks 18 and 19 from direct discard is expected to be negligible. Any Th-232 and Ra-228 that result from the decay of Pu-240 or U-236 is accounted for by the performance assessment model.

The predicted inventories of Ra-226 and Th-230 are based on their decay from the sample-derived inventory of U-234. The methodology for performing these calculations is described in CBU-

PIT-2005-00040.

The predicted inventory of Th-229 is based on its decay from the sample-derived inventory of U-233. The methodology for performing this calculation is described in CBU-PIT-2005-00040.

The predicted inventories of Ac-227 and Pa-231 are based on their decay from the sample-derived inventory of U-235. The methodology for performing these calculations is described in CBU-PIT-2005-00040.

Overall, while there is some uncertainty in predicted heel inventories, it can be seen by comparison to sampled inventories that predicted inventories that are based on fission-yield calculations are most likely over-predicted and that predicted inventories based on accountability records could possibly be under-predicted. The uncertainty in accountability record predictions is not impactive, however, since the inventories used for the affected radionuclides were derived from sample results.

**Additional discussion of specific examples in the NRC's Basis for the Comment**

*The sampled values for uranium isotopes range from a factor of four to 30 less in Tank 19 compared to Tank 18; however, the WCS generated value for U-232 is nearly identical.*

Unlike other uranium isotopes whose inventories in the waste tanks are based on accountability records, U-232 is characterized by the fission-yield computer code methodology. No credit is taken for removal of U-232 during canyon processing and, for PUREX waste, 36% is discarded to the low activity stream. Since the characterization basis for U-232 is different from other uranium isotopes, the ratio of U-232 to other uranium isotopes is expected to vary from tank to tank.

*The Cm-244 value in Tank 19 generated with the WCS is roughly five orders of magnitude less than the value for Tank 18, even though the other Cm isotopes are comparable.*

See discussion of Cm-244.

*The Pu-244 value (from special calculation) in Tank 19 is roughly four times higher than the value for Tank 18; however, all the sampled isotopes of Pu are less in Tank 19 than Tank 18.*

As discussed above, the Pu-244 inventory is based on the assumption that the mass of Pu-244 in a tank is equal to the mass of Pu-242 in the tank. The mass of Pu-242 in Tank 19 (based on sample results) is roughly four times higher than the mass of Pu-242

in Tank 18 (based on sample results); therefore, the Pu-244 inventory in Tank 19 is predicted to be roughly four times higher than the Pu-244 inventory in Tank 18.

*The Characterization of Tank 19 Residual Waste [15] indicates that, prior to sampling, the concentration of Cs-137 in Tank 19 solids was under-predicted by factor of approximately 200. The under-prediction is discussed and attributed to partitioning of Cs-137 onto zeolite. However, it is not clear that Cs-137 is the only radionuclide that is likely to partition onto the zeolite, and the possible under-prediction of the concentration of unsampled radionuclides in Tank 19 solids due to partitioning onto zeolite is not discussed.*

See discussion regarding zeolite.

*The Characterization of Tank 19 Residual Waste [15] indicates that, prior to sampling the concentration of Pu-242 in the Tank 19 solids was under-predicted by approximately a factor of 10. No reason for the under-prediction is discussed, and no assessment of the possibility that unsampled radionuclides could be under-predicted by a similar factor due to similar processes is provided.*

The sample-derived inventory for Tank 19 is based on less-than-detection sample results; therefore, no conclusions can be drawn regarding the accuracy or inaccuracy of the Tank 19 Pu-242 inventory prediction. The predicted Pu-242 inventory is based on accountability records and, as a result, cannot be used to evaluate the potential for unsampled radionuclides to be under-predicted since none of the unsampled radionuclides inventories are based on accountability records.

*Tanks 17 and 19 are estimated (based on sampling) to contain the two highest Tc-99 concentrations in the tank farm. It is unlikely that the Tc-99 inventory in all of the F Tank Farm tanks that have not been sampled is lower than these sampled values, unless there is a reason to expect that Tanks 17 and 19 contain the waste with the highest Tc-99 concentrations in the tank farm. Thus, it seems that the Tc-99 inventories in the other tanks in the tank farm have been under-estimated.*

CBU-PIT-2005-00140 provides an estimate of the inventory of Tc-99 in the residual material in the tanks at the time of closure. The inventory predicted for F- Tank Farm tanks at closure is based on radionuclide concentrations taken from the SRS WCS combined with assumptions about the mass of material remaining in each tank. (CBU-PIT-2005-00140, page 6.) The F-Tank Farm tank inventory is based on the mass of 100 gallons of sludge in each tank containing high activity waste and the mass of 1000 gallons of sludge in each tank containing low activity waste. (CBU-PIT-2005-

00140, page 8) Tanks 7, 25, and 27 are the only tanks in the F-Tank Farm that are expected to contain zeolite, so the Tanks 7, 25 and 27 inventories have been adjusted to account for zeolite. (CBU-PIT-2005-00140, page 6) The method for predicting the inventory in the tanks at the time of closure does not credit the potential removal of the more soluble radionuclides during the waste removal and heel removal processes.

The Tc-99 concentration in the other tanks in the tank farm will be characterized through sampling prior to closure.

*The sampled Tank 18 value for Np-237 is by far the highest value of all tanks. DOE acknowledges that part of the reason for the high Np-237 in Tank 18 is the introduction of laboratory waste that was not tracked with the WCS [18]. The result demonstrates that untracked waste can have a significant impact on estimates generated with the WCS.*

The only radionuclides whose inventories can be skewed through "untracked discards" are those that were separated during processing due to their desirability. The inventories of these materials in Tanks 18 and 19 have been characterized through analysis of residual material samples.

*The total inventory of Np-237 in Tanks 17-20 is roughly three times higher than the predicted inventory in Tanks 1-8. However, the concentration of Np-237 in Tanks 17-20 is roughly 30 times lower than the predicted concentration in Tanks 1-8. This suggests that either DOE has assumed roughly 100 times better waste removal for Tanks 1-8 than for Tanks 17-20 when generating the inventory and the overall risk from the F Tank Farm or the concentrations are inaccurate.*

CBU-PIT-2005-00140 provides an estimate of the inventory of Np-237 in the residual material in the tanks at the time of closure. The inventory predicted for F- Tank Farm tanks at closure is based on radionuclide concentrations taken from the SRS WCS combined with assumptions about the mass of material remaining in each tank. (CBU-PIT-2005-00140, page 6.) The F-Tank Farm Tanks 1-8 inventory is based on the mass of 100 gallons of sludge in each tank. (CBU-PIT-2005-00140, page 8) Tank 7 is the only tank in the Tanks 1-8 "eight-pack" that is expected to contain zeolite, so the Tank 7 inventory has been adjusted to account for zeolite. (CBU-PIT-2005-00140, page 6) The method for predicting the inventory in the tanks at the time of closure does not credit the potential removal of the more soluble radionuclides during the waste removal and heel removal processes.

*There is zero estimated tritium in Tanks 1-8 and Tanks 25-28, 33-*

*34, and 44-47, although the sampled tank group (Tanks 17-20) has measured tritium.*

The tritium in Tanks 17-20 was found in the residual liquid samples. For the other tanks in the tank farms, it is assumed that there will not be a significant contribution from residual liquids. At the completion of bulk waste removal and the planned two phases of heel removal, the predicted small volume of residual liquid is assumed to have minimal concentrations of radionuclides following mechanical mixing and chemical treatment.

*The concentration of I-129 is lowest for the tank group of 17-20, as estimated with the WCS, compared to the other tank groups. It is not clear why I-129 concentrations are so much lower for Tanks 17-20 than the other tanks.*

There are two reasons for the low I-129 concentrations in Tanks 17-20. First, 95% of the I-129 discarded to waste was discarded with the high heat waste stream. The other 5% was discarded with the LHW stream. As a result, low heat tanks like Tanks 17-20 are nominally a factor of 20 times lower than high heat tanks. In addition to this, since Tanks 17-20 were single-shell uncooled tanks, they received a disproportionate amount of lower fission product bearing waste streams such as cladding waste. For example, 31% (by mass) of the transfers into Tank 18 contained negligible fission products. As a result of these two factors, the fission product concentrations in Tanks 17-20 are expected to be low relative to the other tanks.

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**Table 15-1: Comparison of All Solids Predictions to Sample Results**

| Tank | Nuclide | Prediction Method | Solids (Ci) | | Sample to Prediction Ratio |
|---|---|---|---|---|---|
| | | | Predicted*** | Sample (Average) | |
| 18 | Se-79 | Fission Yield | 7.54E-02 | 2.21E-02 | 0.29 |
| 18 | Sr-90 | Fission Yield | 3.41E+03 | 1.16E+03 | 0.34 |
| 18 | Y-90 | Fission Yield | 3.41E+03 | 1.16E+03 | 0.34 |
| 18 | Tc-99 | Fission Yield | 1.77E+01 | 1.35E+00 | 0.08 |
| 18 | Cs-137 | Fission Yield | 2.38E+02 | 9.61E+03 | 40 |
| 18 | Ba-137m | Fission Yield | 2.25E+02 | 9.09E+03 | 40 |
| 18 | U-235 | Accountability | 1.66E-03 | 6.60E-03 | 3.97 |
| 18 | U-238 | Accountability | 8.95E-02 | 1.70E-01 | 1.90 |
| 18 | Pu-238 | Accountability | 3.81E+02 | 6.12E+01 | 0.16 |
| 18 | Pu-239 | Accountability | 5.44E+01 | 1.24E+02 | 2.27 |
| 18 | Pu-240 | Accountability | 1.39E+01 | 2.75E+01 | 1.98 |
| 18 | Pu-241 | Accountability | 2.44E+02 | 2.14E+02 | 0.88 |
| 18 | Pu-242 | Accountability | 1.78E-02 | 5.63E-02 | 3.16 |
| 18 | Am-241 | Accountability* | 2.85E+01 | 6.54E+01 | 2.29 |
| 19 | Se-79 | Fission Yield | 6.83E-02 | 3.60E-02 | 0.53 |
| 19 | Sr-90 | Fission Yield | 3.59E+03 | 3.63E+01 | 0.01 |
| 19 | Y-90 | Fission Yield | 3.59E+03 | 3.63E+01 | 0.01 |
| 19 | Tc-99 | Fission Yield | 1.60E+01 | 5.23E+00 | 0.33 |
| 19 | Cs-137 | Fission Yield | 2.49E+02 | 4.54E+04 | 182 |
| 19 | Ba-137m | Fission Yield | 2.35E+02 | 4.30E+04 | 182 |
| 19 | U-235 | Accountability | 1.37E-03 | 9.37E-04 | 0.68 |
| 19 | U-238 | Accountability | 1.25E-01 | 3.14E-02 | 0.25 |
| 19 | Pu-238 | Accountability | 5.14E+02 | 1.68E+01 | 0.03 |
| 19 | Pu-239 | Accountability | 7.34E+01 | 2.21E+01 | 0.30 |
| 19 | Pu-240 | Accountability | 1.64E+01 | 7.68E+00 | 0.47 |
| 19 | Pu-241 | Accountability | 2.20E+03 | 4.67E+01 | 0.02 |
| 19 | Pu-242 | Accountability | 3.38E-02 | 1.63E-01 | 4.83 |
| 19 | Am-241 | Accountability** | 2.04E+02 | 7.13E+00 | 0.03 |

*97% of the predicted Tank 18 Am-241 inventory is from decay of Pu-241

**100% of the predicted Tank 19 Am-241 inventory is from decay of Pu-241

***From CBU-PIT-2005-00067 for Tank 18 and CBU-PIT-2005-00068 for Tank 19

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**Table 15-2a: Comparison of Fission Yield Predictions to Unpublished Gamma Scan Results for Tank 19**

| Isotope | Concentration in solids (uCi/g) | Sample Based Solids Inventory (Ci) | Predicted Solids* Inventory (Ci) | Sample to Prediction Ratio | Comments |
|---|---|---|---|---|---|
| Co-60 | 8.42E-03 | 4.64E-01 | 7.22E+00 | 6.4E-02 | (Actual concentration value) |
| Nb-94 | <4.13E-04 | <2.28E-02 | 3.62E-05 | <6.3E+02 | |
| Ru-106 | <6.6E-03 | <3.64E-01 | 2.70E-04 | <1.3E+03 | |
| Rh-106 | <6.6E-03 | <3.64E-01 | 2.70E-04 | <1.3E+03 | (determined from Ru-106, same as Ru-106) |
| Sn-126 | <4.34E-04 | <2.39E-02 | 1.27E-01 | <1.9E-01 | |
| Sb-125 | <3.04E-03 | <1.68E-01 | 2.59E+00 | <6.5E-02 | |
| Ce-144 | <3.24E-03 | <1.79E-01 | 3.71E-06 | <4.8E+04 | |
| Eu-152 | <2.66E-03 | <1.47E-01 | 5.62E-03 | <2.6E+01 | |
| Eu-154 | <8.92E-04 | <4.92E-02 | 1.47E+01 | <3.3E-03 | |
| Eu-155 | <1.41E-03 | <7.77E-02 | 7.57E-02 | <1.0E-00 | |
| Ra-226 | <1.49E-02 | <8.21E-01 | 1.06E-07 | <7.8E+06 | |
| Cm-243 | <4.07E-03 | <2.24E-01 | 2.70E-06 | <8.3E+04 | |
| Cm-245 | <1.39E-03 | <7.66E-02 | 1.96E-09 | <3.9E+07 | |
| Cf-249 | <1.17E-03 | <6.45E-02 | 3.96E-22 | <1.6E+20 | |

*Predicted values from CBU-PIT-2005-00068

Attachment 1

**Table 15-2b: Comparison of Fission Yield Predictions to Unpublished Gamma Scan Results for Tank 18**

| Isotope | Average of samples FTF-213, -214, -216, -228, and -229 | Remainder Inventory (in 13,131 kg) | Sample FTF-230 | Mound Inventory (in 2044 kg) | Total Sample-based Inventory | Predicted (CBU-PIT-2005-00067) | Sample to Predicted Ratio |
|---|---|---|---|---|---|---|---|
| | uCi/g | Ci | uCi/g | Ci | Ci | Ci | |
| Co-60 | 8.53E-02 | 1.12E+00 | 6.18E-02 | 1.26E-01 | 1.25E+00 | 4.32E+00 | 2.9E-01 |
| Nb-94 | <7.88E-03 | <1.04E-01 | <7.21E-03 | <1.47E-02 | <1.18E-01 | 4.00E-05 | <3.0E+03 |
| Ru-106 | <4.63E-02 | <6.08E-01 | <4.19E-02 | <8.57E-02 | <6.94E-01 | 9.72E-05 | <7.1E+03 |
| Rh-106 | <4.63E-02 | <6.08E-01 | <4.19E-02 | <8.57E-02 | <6.94E-01 | 9.72E-05 | <7.1E+03 |
| Sn-126 | <2.40E-02 | <3.15E-01 | <2.05E-02 | <4.18E-02 | <3.57E-01 | 1.40E-01 | <2.6E+00 |
| Sb-125 | <2.38E-02 | <3.13E-01 | <2.26E-02 | <4.61E-02 | <3.59E-01 | 1.17E+00 | <3.1E-01 |
| Ce-144 | <7.18E-02 | <9.43E-01 | <6.08E-02 | <1.24E-01 | <1.07E+00 | 1.44E-06 | <7.4E+05 |
| Eu-152 | <2.58E-02 | <3.39E-01 | <2.41E-02 | <4.93E-02 | <3.88E-01 | 1.98E-01 | <2.0E+00 |
| Eu-154 | 4.00E-02 | 5.25E-01 | 2.65E-02 | 5.41E-02 | 5.79E-01 | 1.06E+01 | 5.5E-02 |
| Eu-155 | <2.37E-02 | <3.12E-01 | <1.71E-02 | <3.49E-02 | <3.47E-01 | 2.67E+00 | <1.3E-01 |
| Ra-226 | <2.01E-01 | <2.64E+00 | <1.78E-01 | <3.65E-01 | <3.01E+00 | 4.90E-07 | <6.1E+06 |
| Cm-243 | <3.59E-02 | <4.72E-01 | <3.11E-02 | <6.36E-02 | <5.35E-01 | 9.52E-05 | <5.6E+03 |
| Cm-245 | <7.82E-02 | <1.03E+00 | <6.69E-02 | <1.37E-01 | <1.16E+00 | 2.17E-09 | <5.4E+08 |
| Cf-249 | <1.07E-02 | <1.40E-01 | <9.74E-03 | <1.99E-02 | <1.60E-01 | 1.39E-20 | <1.2E+19 |

**Table 15-3: Comparison of Fission Yield Predictions to Sample Results**

| Tank | Nuclide | Prediction Method | Solids (Ci) | | Sample to Prediction Ratio |
|---|---|---|---|---|---|
| | | | Predicted | Sample-Based | |
| 18 | Se-79 | Fission Yield | 7.54E-02 | 2.21E-02 | 0.29 |
| 18 | Sr-90 | Fission Yield | 3.41E+03 | 1.16E+03 | 0.34 |
| 18 | Y-90 | Fission Yield | 3.41E+03 | 1.16E+03 | 0.34 |
| 18 | Tc-99 | Fission Yield | 1.77E+01 | 1.35E+00 | 0.08 |
| 18 | Cs-137 | Fission Yield | 2.38E+02 | 9.61E+03 | 40 |
| 18 | Ba-137m | Fission Yield | 2.25E+02 | 9.09E+03 | 40 |
| 19 | Se-79 | Fission Yield | 6.83E-02 | 3.60E-02 | 0.53 |
| 19 | Sr-90 | Fission Yield | 3.59E+03 | 3.63E+01 | 0.01 |
| 19 | Y-90 | Fission Yield | 3.59E+03 | 3.63E+01 | 0.01 |
| 19 | Tc-99 | Fission Yield | 1.60E+01 | 5.23E+00 | 0.33 |
| 19 | Cs-137 | Fission Yield | 2.49E+02 | 4.54E+04 | 182 |
| 19 | Ba-137m | Fission Yield | 2.35E+02 | 4.30E+04 | 182 |
| 19 | Co-60 | Fission Yield | 7.22E+00 | 4.64E-01 | 0.06 |
| 19 | Nb-94 | Fission Yield | 3.62E-05 | <2.28E-02 | <6.3E+02 |
| 19 | Ru-106 | Fission Yield | 2.70E-04 | <3.64E-01 | <1.3E+03 |
| 19 | Rh-106 | Fission Yield | 2.70E-04 | <3.64E-01 | <1.3E+03 |
| 19 | Sn-126 | Fission Yield | 1.27E-01 | <2.39E-02 | <0.19 |
| 19 | Sb-125 | Fission Yield | 2.59E+00 | <1.68E-01 | <0.06 |
| 19 | Ce-144 | Fission Yield | 3.71E-06 | <1.79E-01 | <4.8E+04 |
| 19 | Eu-152 | Fission Yield | 5.62E-03 | <1.47E-01 | <2.6E+01 |
| 19 | Eu-154 | Fission Yield | 1.47E+01 | <4.92E-02 | <0.003 |
| 19 | Eu-155 | Fission Yield | 7.57E-02 | <7.77E-02 | <1.0 |
| 19 | Cm-243 | Fission Yield | 2.70E-06 | <2.24E-01 | <8.3E+04 |
| 19 | Cm-245 | Fission Yield | 1.96E-09 | <7.66E-02 | <3.9E+07 |
| 19 | Cf-249 | Fission Yield | 3.96E-22 | <6.45E-02 | <1.6E+20 |
| 18 | Co-60 | Fission Yield | 4.32E+00 | 1.25E+00 | 2.9E-01 |
| 18 | Nb-94 | Fission Yield | 4.00E-05 | <1.18E-01 | <3.0E+03 |
| 18 | Ru-106 | Fission Yield | 9.72E-05 | <6.94E-01 | <7.1E+03 |
| 18 | Rh-106 | Fission Yield | 9.72E-05 | <6.94E-01 | <7.1E+03 |
| 18 | Sn-126 | Fission Yield | 1.40E-01 | <3.57E-01 | <2.6E+00 |
| 18 | Sb-125 | Fission Yield | 1.17E+00 | <3.59E-01 | <3.1E-01 |
| 18 | Ce-144 | Fission Yield | 1.44E-06 | <1.07E+00 | <7.4E+05 |
| 18 | Eu-152 | Fission Yield | 1.98E-01 | <3.88E-01 | <2.0E+00 |
| 18 | Eu-154 | Fission Yield | 1.06E+01 | 5.79E-01 | 5.5E-02 |
| 18 | Eu-155 | Fission Yield | 2.67E+00 | <3.47E-01 | <1.3E-01 |
| 18 | Cm-243 | Fission Yield | 9.52E-05 | <5.35E-01 | <5.6E+03 |
| 18 | Cm-245 | Fission Yield | 2.17E-09 | <1.16E+00 | <5.4E+08 |
| 18 | Cf-249 | Fission Yield | 1.39E-20 | <1.60E-01 | <1.2E+19 |

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**Table 15-4: Comparison of Tank 19 Solids Samples**

| Species | Mound Sample (WSRC-RP-97-0074) (µCi/g) | Crusty Solids (WSRC-RP-2001-00410) (µCi/g) | Residual Material Sample Average (CBU-PIT-2005-00068) (µCi/g) |
|---|---|---|---|
| Sr-90 | 1.69E+00 | 5.30E-01 | 6.58E-01 |
| Tc-99 | 1.2E-01 | 2.05E-02 | 9.49E-02 |
| Cs-137 | 9.08E+02 | 6.28E+01 | 8.25E+02 |
| Np-237 | 3.0E-04 | 1.98E-04 | 1.61E-04 |
| U-233 | <Detection Limit (DL) | 6.28E-04 | <DL |
| U-235 | 1.1E-05 | 8.56E-06 | 1.70E-05 |
| U-238 | 3.6E-04 | 2.80E-04 | 5.70E-04 |
| Pu-238 | 3.1E+00 | 5.05E-01 | 3.05E-01 |
| Pu-239 | 1.6E-01 | 1.55E-01 | 4.01E-01 |
| Pu-240 | 8.0E-02 | 5.75E-02 | 1.39E-01 |
| Pu-242 | NA* | 2.88E-04 | <DL |

*NA: not analyzed/reported

**Table 15-5: Americium-242m Supernate Sample Results (X-ESR-G-00004, p. 11)**

| Tank | Am-242m (pCi/ml) |
|---|---|
| 13 | <1.15E+04 |
| 30 | <2.72E+03 |
| 37 | <7.22E+03 |
| 39 | <3.70E+03 |
| 45 | <1.87E+03 |
| 49 | <1.82E+05 |

**Table 15-6: Carbon-14 Supernate Sample Results (X-ESR-G-00004, p. 11)**

| Tank | C-14 (pCi/ml) |
|---|---|
| 2 | <1.22E+03 |
| 3 | <2.20E+03 |
| 13 | <4.51E+02 |
| 30 | <7.56E+02 |
| 37 | 2.69E+02 |
| 39 | 1.71E+03 |
| 46 | <7.11E+03 |
| 49 | <3.43E+02 |

**Table 15-7: Fission Yield Activity ratios, 30 yr Aged Waste (CBU-PIT-2005-00068)**

| Activity Ratio | |
|---|---|
| Sm-151 /Sr-90 | 3.25E-02 |
| Eu-152 /Sr-90 | 1.41E-04 |
| Eu-155 /Sr-90 | 1.89E-03 |
| Am-243 /Sr-90 | 4.37E-09 |
| Cm-242 /Sr-90 | 2.27E-23 |
| Cm-243 /Sr-90 | 6.76E-08 |
| Cm-247 /Sr-90 | 1.54E-21 |
| Cm-248 /Sr-90 | 3.56E-22 |
| Bk-249 /Sr-90 | 1.34E-31 |
| Cf-249 /Sr-90 | 9.90E-24 |