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# Los Alamos
NATIONAL LABORATORY

# Memorandum

**TRU Waste Characterization (RRES-CH)**

**Risk Reduction and Environmental Stewardship Division**

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**Subject:** $^{234}$U and $^{90}$Sr Calculations For NDA Reporting

## References

1. Smith, C., *Documentation of Reconciliation of Non-Destructive Assay Isotopic Measurement Data with Acceptable Knowledge information using Microsoft Access 2000 database "NDA-AK Reconciliation v1.0.mdb"*, Los Alamos National Laboratory Memorandum, TWCP-10108, October 9, 2002.
2. Baker, M., *Assay of U-234*, Los Alamos National Laboratory Memorandum, TWCP-09810, September 12, 2002.
3. Miller, G., *Estimation of $^{234}$U from $^{238}$Pu Decay*, Los Alamos National Laboratory Memorandum, TWCP-10446, November 5, 2002.
4. TWCP-AK-2.1-015, R.2, *Acceptable Knowledge Report for Legacy Debris TA-55 Waste Streams Containing Pu-239*, Los Alamos National Laboratory, Report LA-UR-02-6665, October 9, 2002.
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The uranium isotope $^{234}$U and the strontium isotope $^{90}$Sr have been reported via the NDA reconciliation report (Ref. 1) and it is desired to report the data as part of the NDA assay. $^{234}$U has extremely weak gamma energies in the region used by the gamma system (FRAM) that would require kilogram amounts to be detectable. $^{90}$Sr produces no gamma ray and consequently is not detectable by the current NDA systems. This memorandum outlines the method to be implemented in the NDA reports to account for these two isotopes.

## $^{234}$U

The present approach used in the FRAM NDA system assesses the region around 454.92 keV for the presence of $^{234}$U rather than the more intense gamma ray at 120.91 keV which is subject to too much interference to be useable (Ref. 2). Unfortunately, the 454.92 keV gammas have a branching ratio of only 2.6 x $10^{-7}$ and are not suitable for detection of $^{234}$U at sub-kilogram levels. This check will continue to be made in FRAM but an alternative method for determining lower quantities is needed.

A second approach to quantification utilizes the fact that the daughter of $^{238}$Pu is $^{234}$U by alpha decay. The $^{238}$Pu gamma spectrum is quantifiable by FRAM. After 45 years of decay (a time period providing a conservative mass estimate), the mass, m, and the uncertainty, σ, of $^{234}$U become (Ref. 3):

$$m_{U234} = 0.427 m_{Pu238} \qquad \sigma_{U234} = 0.427\sigma_{Pu238} \qquad (1)$$

The third approach to quantifying $^{234}$U considers the production of uranium material. The most conservative ratio of mass abundance of $^{234}$U and $^{235}$U has been found to be 0.014 (Ref. 4). The gamma spectrum from $^{235}$U is quantifiable by FRAM. Consequently, for uranium production, the mass and uncertainty of $^{234}$U may be obtained from the equation:

$$m_{U234} = 0.014 m_{U235} \qquad \sigma_{U234} = 0.014 \sigma_{U235} \qquad (2)$$

In implementing the assay, the mass value reported for $^{234}U$ will be derived from the maximum value reported by FRAM or the sum of the masses from the $^{238}Pu$ and $^{235}U$ correlations, as shown in equations ( 3) and ( 4).

$$m_{U234} = MAX\{m_{FRAM}, (0.427 m_{Pu238} + 0.014 m_{U235})\} \qquad (3)$$

$$\sigma_{U234} = MAX\{\sigma_{FRAM}, [\sqrt{(0.427\sigma_{Pu238})^2 + (0.014\sigma_{U235})^2}]\} \qquad (4)$$

## $^{90}Sr$

Strontium is a byproduct of the fission process, as is $^{137}Cs$. While $^{137}Cs$ produces a very strong and quantifiable gamma peak even in microgram quantities, $^{90}Sr$ is a beta particle emitter and produces no gamma signal. The fission product yield has been quantified for several isotopes, including $^{239}Pu$, $^{241}Pu$, and uranium (Ref. 5). Taking ratios of the relative yields from $^{239}Pu$ and equating these to the ratio of the number of atoms of $^{90}Sr$ to $^{137}Cs$, a correlation between the masses is obtained (see appendix). This is given by

$$m_{Sr90} = (0.205 \pm 0.011) m_{Cs137} \qquad \sigma_{Sr90} = m_{Sr90}\sqrt{0.00288 \pm \left(\frac{\sigma_{Cs137}}{m_{Cs137}}\right)^2} \qquad (5)$$

Current procedures (Ref. 4) make use of the assertion that the activities of the two isotopes are approximately equal, in which case the above equation becomes

$$m_{Sr90} = 0.636 m_{Cs137} \text{ (Equal Activities)} \qquad \sigma_{Sr90} = 0.636 \sigma_{Cs137} \qquad (6)$$

Clearly, applying equal activities overstates the mass of $^{90}Sr$ by a factor of 3.1 but produces a conservative estimate of $^{90}Sr$. Since $^{137}Cs$ has not been observed approaching 5% of the total activity, continuing the use of equality of activities will not seriously bias the $^{90}Sr$ assay. Consequently, in the assay spreadsheet, $^{90}Sr$ will be reported at the quantity expressed by equation ( 6) for equality of the activities of $^{90}Sr$ and $^{137}Cs$.

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1 Attachment: Appendix

Technical Review completed.

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APPENDIX

## Definitions

| | |
|---|---|
| $\eta$ | Number of moles |
| M | Molecular Weight (g/mole) (Ref. 6)<br>Sr90 = 89.908 g/mole; Cs137 = 136.907 g/mole |
| m | Mass (g) |
| $\sigma$ | Mass uncertainty (g) |
| N | Number of molecules |
| $N_A$ | Avogadro's Number (molecules/mole) |
| A | Activity (Bq) |
| k | Subscript representing an isotope |
| t | Half-life (Ref. 6)<br>Sr90 = 29.1 y; Cs137 = 30.3 y |
| $Y_k$ | Yield of isotope k with respect to Nd for Pu239 (Ref. 5)<br>Sr90 = 1.233 +/- 0.007; Cs137 = 3.970 +/- 0.014 |
| F | Number of fissions |

## Mass based on Pu239 fission yields

$$\eta_k = \frac{m_k}{M_k} = \frac{N_k}{N_A} \qquad (7)$$

$$N_k = Y_k Y_{Nd} F \qquad (8)$$

$$A_k = \frac{N_k \ln(2)}{t_k} \qquad (9)$$

$$\frac{\eta_{Sr90}}{\eta_{Cs137}} = \frac{m_{Sr90} M_{Cs137}}{m_{Cs137} M_{Sr90}} = \frac{N_{Sr90}}{N_{Cs137}} = \frac{Y_{Sr90}}{Y_{Cs137}} = \frac{A_{Sr90}}{A_{Cs137}} \frac{t_{Sr90}}{t_{Cs137}} \qquad (10)$$

$$m_{Sr90} = \frac{M_{Sr90}}{M_{Cs137}} \frac{Y_{Sr90}}{Y_{Cs137}} m_{Cs137} = 0.204 m_{Cs137} \qquad (11)$$

$$\sigma_{Sr90} = m_{Sr90} \sqrt{(0.007)^2 + \left(\frac{\sigma_{Cs137}}{m_{Cs137}}\right)^2} \qquad (12)$$

## Mass based on equal activity ( $A_{Sr90} = A_{Cs137}$ )

$$m_{Sr90} = \frac{M_{Sr90}}{M_{Cs137}} \frac{t_{Sr90}}{t_{Cs137}} m_{Cs137} = 0.631 m_{Cs137} \qquad (13)$$

$$\sigma_{Sr90} = 0.631 \sigma_{Cs137} \qquad (14)$$