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Traditional and Model Based Assay of Irregular Geometry Items

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ABSTRACT

The Analytical Development Section (ADS) of SRNL was requested to perform a waste disposal assay of two heater boxes which had been used in the HB Line dissolvers. They had been sent to SRNL for study to make recommendations on how to prevent future failure of the units when they were replaced. The study having been completed, the units needed to be characterized prior to sending to Solid Waste for disposal. An assay station consisting of a turntable, HPGe detector, CANBERRA Inspector, transmission source and a portable computer was set up to do the required assays. The assays indicate the presence of U-235, Pu-239 and Cs-137. No measurable amounts of U-235 or Pu-239 were found. Therefore the Minimum Detectable Activities for U-235 and Pu-239 were calculated. For Heater Box 1, 0.23 grams of U-235 and 0.24 grams of Pu-239. For Heater Box 2, the results were 0.21 grams of U-235 and 0.21 grams of Pu-239. This paper describes and documents the assays employed to determine the amount of U, Pu and Cs contents of the heater boxes. The paper provides results of SNM assays using traditional calibration of the system and on one based on modeling. It also provides the scientific community with data that will assist the user in determining the method of choice for assaying items with irregular geometries.

SUMMARY

The Analytical Development Section of SRNL was requested to assay two Heater Boxes for U-235 and Pu-239 contamination. The boxes were used in the HB Line dissolvers. The boxes had been studied to learn failure modes. Upon completion of the study the boxes had to be assayed to determine the proper waste stream for disposal. The boxes were assayed in a temporary assay facility set up in room F090 in building 773A. Accountability data gave estimates of 0.3 gm Pu and 0.6 gm enriched U for one box and 0.2 grams Pu and 0.2 grams enriched U for the other. The assay was requested to obtain values for waste disposal purposes. Table 1 gives the results of the assay. The methodology and results of the analysis are discussed below.

	U-235 grams	Pu-239 grams	Cs-137 microCi
Heater Box 1	<0.23	<0.24	<0.44
Heater box 2	<0.21	<0.21	<0.15

Table 1 Measurement results

METHODOLOGY

A temporary assay facility was set up in room F090 in building 773A. The assay facility consisted of a turntable to rotate the heater boxes, and a High Purity Germanium (HPGE) detector and associated electronics. Rotation of the sample ensures that inhomogenities within the sample do not distort the results. A Cs-137 transmission source was set up

behind the heater box to permit compensation for attenuation of the gamma rays by the material of the heater box and its contents. Data was acquired using a Canberra Inspector™ controlled by a laptop computer containing the Canberra GENIE2K™ (Reference 1) analysis software package. The two heater boxes were metal boxes 12" by 12" by 48" containing heating coils and individually wrapped in plastic for contamination control. Each heater box was stood vertically on the turntable and the HPGE detector was located 60 inches from the edge of the box at an elevation 24 inches from the base of the heater box. Figure 1 shows the assay facility, the turntable with the heater box mounted on it, and the HPGE detector mounted on its transport cart.



Figure 1 Temporary Assay Facility with wrapped heater box on turntable.

A thirty minute room background count was acquired before the measurements were begun, and again after the measurements were completed. The final background count was stripped from the beginning background. The resulting spectrum was statistically small, demonstrating that conditions had not changed during the measurement time. The initial background measurement is shown in Figure 2.

A one hour data acquisition was made of each heater box. Since there was no identification of the specific heater box written on the plastic wrapper, the first heater box assayed was denoted "Heater Box 1" (HB1) and the second "Heater Box 2" (HB2). After the data for HB1 had been acquired, a Cs-137 source was obtained and placed behind the heater box, and a fifteen minute count of the Cs-137 gamma rays passing through the box was obtained. This transmission measurement was denoted as "XM1". The spectrum obtained in the Heater Box 2 assay is shown in Figure 3. The Uranium-235, Pu-239 and Cs-137 gamma ray regions are indicated. The Annihilation radiation location is denoted because it is present in all the spectra and can be used as an energy calibration value.

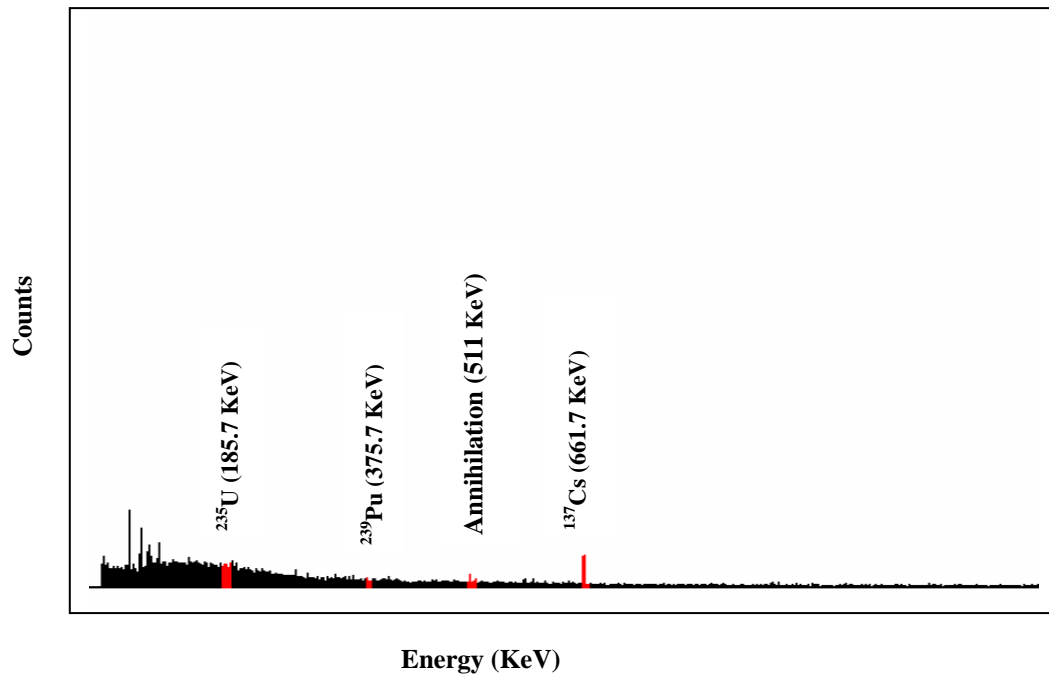


Figure 2, Background 1.

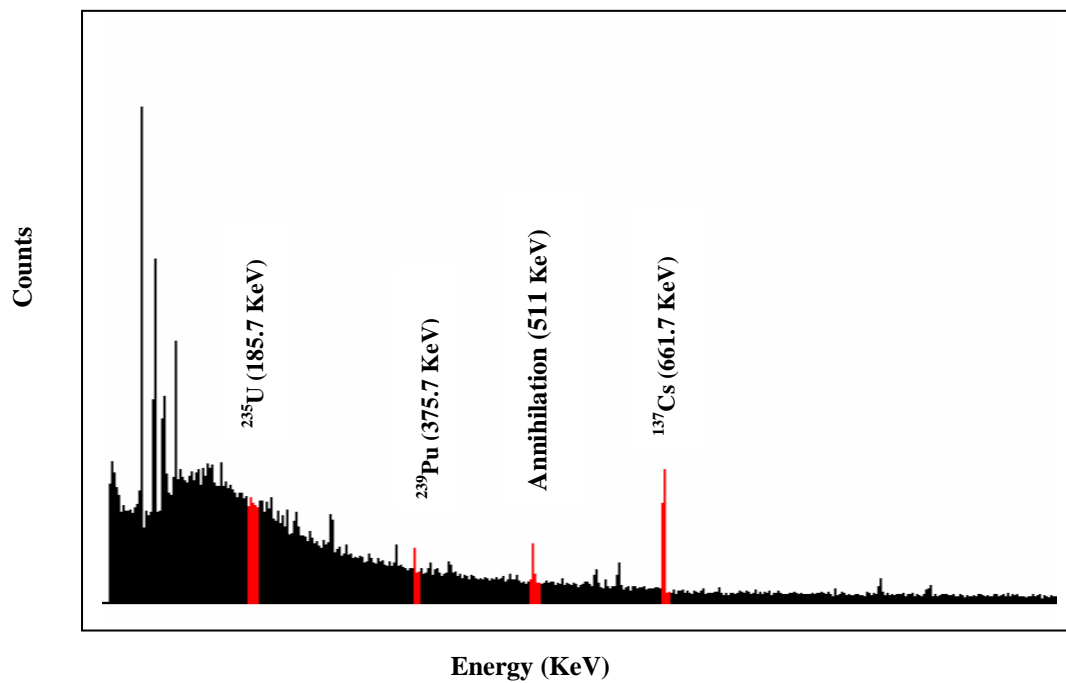


Figure 3, Heater Box 2 one hour acquisition

Heater Box 1 was removed and a 15 minute count of the Cs-137 source alone was obtained and denoted "T0". Any contamination of the heater boxes would come from contamination on the inside of the box and gammas from the contamination would be attenuated by the box wall. From the two counts, T0 and XM1, an estimate of the attenuation could be obtained.

For Heater Box 2, the process was repeated; a transmission measurement “XM2” and a one hour assay HB 2 of the erect heater box were obtained. This was followed by the final background acquisition.

Any contamination of the heater boxes will be on the inside of the boxes. These gamma rays will be attenuated when passing through the steel wall of the heater box and hence result in an underestimation of the contamination. A transmission correction is used to compensate for this effect (Reference 2). A transmission source (in this case Cs-137) is placed behind the heater box and a count made of the Cs-137 (0.662 MeV) gamma rays passing through the heater box. A count is also made with the object removed, in which case the detector sees only the transmission source. As can be seen from Table 1, there is also Cs-137 in the room background. This must be subtracted from each of the data points. The average Cs-137 count in the two background runs is 0.285c/s.

Therefore, for HB 1, the transmission is $(.44 - .285)/(.91 - .285) = .25$.

Gammas from the transmission source pass through both sides of the heater box, while those within it pass through only one wall. Hence the true transmission is the square root of the above value. The transmission correction is the reciprocal of the transmission or $X_m = 2.0$ for HB 1. Likewise for HB 2 the transmission correction X_m is 1.64.

ANALYSIS

Following data acquisition, the spectra HB 1 and HB 2 were visually studied for evidence of the principal U-235 (185 KeV) and Pu-239 (375 KeV) peaks. No obvious peaks could be seen. Table 2, a summary of the data acquired in the measurements, shows that the count rates in the Pu-239 (375 KeV) and U-235 (185 KeV) peaks, shows that the count rates were very small. The analysis suite of programs in the Genie2K™ program package was employed to locate, energy calibrate, and compute the area of all the peaks in each spectrum. The efficiency of the detector (Reference 3) was an input parameter to correct the area of the peaks for the detector’s varying efficiency with energy. The results of this analysis were input into the ORTEC program ISOTOPIC™. (Reference 4)

Acquisition	185 KeV (cps)	375 KeV (cps)	662 KeV (cps)
BKG 1	.058 +/- 35%	.022 +/- 74	0.25 +/- 7%
Heater box 1	0.014 +/- 119%	.054 +/- 21%	0.366 +/- 4%
Heater box 2	0.018 +/- 88%	.058 +/- 18%	0.313 +/- 4%
XM 1	N/A	N/A	0.44 +/- 8%
XM 2	N/A	N/A	0.52 +/- 7%
T0	N/A	N/A	0.91 +/- 5%
BKG 2	0	.066 +/- 43%	0.32 +/- 10%

Table 2 measured count rates.

A module in the ISOTOPIC™ program allows one to define the geometry of the object being evaluated. The dimensions, mass, distance from the detector of the sample and the specifics of the detector collimator are input values. After defining the heater box, ISOTOPIC™ was executed to obtain the amounts of U-235 or Pu-239 in the heater box. No measurable U-235 or Pu-239 was detected by ISOTOPIC™.

If no activity can be measured, the activity is less than the minimum detectable activity (MDA) for the experimental setup. It is then necessary to compute the MDA for each heater box for each of the isotopes of interest

Uranium-235

At 60 inches from the detector, the heater box is a line source. From the LANL Training program (Reference 5)

$$\text{grams} = \text{Cr} * K_1 * r * X_m * L;$$

where: Cr is measured count rate

K_1 is the line source calibration factor

r is the distance from the detector to the object

X_m is the transmission correction

L is the length of the object.

For Uranium K_1 was obtained from Salaymeh and Dewberry (Reference 6) in which the K_1 was measured for the specific detector used in these measurements and found to be $1.72\text{e-}5 \text{ gm sec/cm}^2$. Table 3 gives the measurement data.

For MDA purposes, the Cr is defined as $3 * \text{sqrt}(\text{bkgnd cr})$ of the gamma ray used in the analysis.

$$r = 60 \text{ in} = 152.4 \text{ cm}$$

$$L = 48 \text{ in} = 121.9 \text{ cm}$$

	MDA CR	XM
Heater Box 1	0.353	2.0
Heater Box 2	0.406	1.64

Table 3 data for Uranium analysis

This yields MDA values of 0.226 grams of U-235 for Heater Box 1 and 0.213 grams for Heater Box 2.

Plutonium-239

For Plutonium-239 there were no values of K_1 available to use in the above method, hence another technique was employed. The assay package in Genie2K™ will compute the MDA at 12 inches (MDA_{12}). ISOTOPIC™ calculates the geometric correction factor from 12 inches to the actual assay distance. Using this value:

$$MDA = MDA_{12} \times GeoCF \times XmCi / gm$$

where: MDA_{12} is obtained from Genie2K™
 GeoCF is the geometric correction factor computed in ISOTOPIC™
 Xm is the transmission correction
 Ci/gm converts the Genie2K™ result from Ci to grams Pu = 0.062 Ci/gm.

For these measurements the geometry correction factor computed in ISOTOPIC™ is GeoCF = 31.74 while the measurement specific data is given in table 4.

	MDA 12	XM
Heater Box 1	2.32e-4 Ci	2.0
Heater Box 2	2.09e-4 Ci	1.64

Table 4 data for Plutonium analysis

This yields MDA values of 0.238 grams for Heater Box 1, and 0.214 grams for Heater Box 2.

Cs-137

It can be seen from the 612 KeV count rates in table 1 that the Cs-137 count rates for Heater Box 1 and Heater Box 2 are significantly above the background level. Therefore there is a small amount of Cs-137 in both boxes. The amount may be estimated as follows:

$$Cs - 137 = \left(\frac{d1}{d2} \right)^2 \times \left(\frac{CRB}{Cr0} \right) \times 4.6 \mu Ci$$

Where: 4.6 μ Ci is the current strength of the transmission source,
 d1 = 60 in, the distance of the heater box from the detector
 d2 = 70 in, the distance of the transmission source from the detector
 CRB is (HB – 0.285), the background corrected Cs count rate from the heater box
 Cr0 is (T0 – 0.285), the background corrected Cs count rate from the transmission source.

This yields 0.44 μ Ci for Heater Box 1 and 0.15 μ Ci for Heater box 2.

DISCUSSION

The U-235 and Pu-239 activity levels in the Heater Boxes are below the Minimum Detectable Activities measurable in the analysis. The MLD values obtained in the measurements are in good agreement with the accountability estimates. The ORTEC ISOTOPIC™ and Canberra Genie2K™ code packages give good results. A combination of these code packages with conventional techniques can be used to obtain good results from items with an irregular geometry in order to obtain data for waste disposal.

5 REFERENCES

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