Gamma-Ray Isotopic Analysis of Plutonium within Highly Attenuating Shipping Containers

Philip A. Hypes, David J. Mercer, Derek R. Dinwiddie
Los Alamos National Laboratory
Los Alamos, NM 87545

Presented at the Institute of Nuclear Materials Management 42nd Annual Meeting Indian Wells, California July 15-19, 2001
Gamma-Ray Isotopic Analysis of Plutonium within Highly Attenuating Shipping Containers

Philip A. Hypes, David J. Mercer, Derek R. Dinwiddie
Los Alamos National Laboratory
Los Alamos, NM 87545

ABSTRACT
Isotopic measurements of items stored in shielded shipping containers presents a challenge to standard, nondestructive high-resolution gamma spectroscopy analysis. For example, some plutonium oxide material that will be shipped from Rocky Flats will be packaged in a combination of containers that places more than 12 mm of lead and 25 mm of steel between the material and the detector. This shielding effectively eliminates gamma rays below approximately 300 keV. Spectra were taken through simulated containers and analyzed using FRAM version 4.0 and a parameter set developed for use with highly attenuated items. The results indicate that 10% precision in measured $^{240}\text{Pu}$ content should be achievable with 2-hour measurements.

INTRODUCTION
It has recently been demonstrated that FRAM [1, 2] software is capable of analyzing high-resolution gamma spectra from plutonium items shielded by as much as 25 mm of lead [3]. This makes FRAM software ideally suited to determine the isotopic composition of plutonium items within highly attenuating shipping containers. Shipping containers, however, often include more than simple lead shielding; they may include layers of lead, steel, and Celotex. Spectra taken under such conditions could be analyzed with the version of FRAM utilized in [3], but the results are significantly improved by analyzing the spectra with the newer version of FRAM, version 4.0 (beta). This version utilizes a physics-based efficiency curve model, which takes into account multiple attenuator matrices such as lead, steel, and cadmium.

MEASUREMENT EQUIPMENT
Measurements were taken with a Canberra coaxial germanium detector, model number GL1015R, with 10% efficiency (relative to a 76 mm diameter x 76 mm long NaI detector). The MCA was a Canberra Inspector 2000, controlled by Genie 2000 software. The shipping containers were simulated with sheets of lead, steel, and Celotex. These attenuating materials were stacked to match the order and thickness of attenuators such as would be encountered by gamma rays produced within highly attenuating shipping containers. The lead layer was 12.5-mm thick. The steel layers totaled 27 mm, and the Celotex layer was 145-mm thick. The source was a 1.5 kg sample of weapons grade (nominally 6% $^{240}\text{Pu}$) PuO$_2$.

MEASUREMENT PROCEDURE
The source and simulated container were positioned above the detector, and a custom MCA control program was initiated to manage data acquisition. All spectra were collected with a gain of 0.125 keV/chn, in 8 k channels. Six measurements were made at each of seven count time values (1000, 2000, 3000, 3600, 4000, 5000, and 6000 seconds).
DATA ANALYSIS
The spectrum in Fig. 1 shows that the lead, steel and Celotex attenuate all plutonium peaks below 320 keV. Because the 160.28 keV $^{240}$Pu peak is unavailable, calculations of the $^{240}$Pu content are based entirely on the 642.48 keV peak; this is the limiting factor in the precision of the measurement. A one-hour measurement provided only approximately 150 counts in this peak, shown in Fig. 2.

![Graph showing the spectrum with peaks at 160.28 keV and 642.48 keV.]

**Fig. 1.** One hour spectrum, showing attenuation of low energy gamma rays.

![Graph showing a detailed view of the 640 keV region with a peak at 642.48 keV.]

**Fig. 2.** Detailed view of 640 keV region, showing sole $^{240}$Pu peak available for analysis.

The spectra were analyzed with FRAM 4.0 (beta), using the physics-based relative efficiency curve model. When using the physics-based relative efficiency curve, the user can specify up to three attenuating materials. Minimum and maximum thickness is specified for each attenuator. The minimum value is usually set to the thickness of the attenuator. The maximum value depends on the distance from the source to the detector, the size of the source, and the size of the detector. Usually, a value about 50% higher than the minimum value is appropriate to account for gamma rays penetrating the attenuator at an oblique angle. Given this information, the software calculates the expected form of the efficiency curve and fits the peaks designated for efficiency curve analysis to the curve.
The parameter set used has been demonstrated to give accurate $^{240}$Pu fractions over a range of isotopic compositions, with up to 25 mm of lead shielding. A FRAM parameter set is a group of user-editable parameters that guides the analysis of a given spectrum. It determines the peaks to be analyzed, the energy calibration, the isotopes of interest, and other parameters. The parameter set structure is responsible for the flexibility and broad utility of the FRAM software.

A typical relative efficiency curve from a lightly shielded item is shown in Fig. 3. The relative efficiency curve for a spectrum taken in the course of this investigation is shown in Fig. 4. This relative efficiency curve was created using the physics-based model. The attenuation parameters entered to create Fig. 4 were 24 to 36 mm of iron, 12.5 to 20 mm of lead, and coaxial type detector.

The analysis results for the experiment are shown in Table I. The “FRAM Uncertainty” values represent the uncertainty calculated by the FRAM software for the mass percentages. The values
in the table are the averages of the six runs taken at each measurement duration (real time). The last column is the relative standard deviation (RSD) of each set of six measurements.

Table I. Analysis results.

<table>
<thead>
<tr>
<th>Count Time(s)</th>
<th>$^{239}$Pu Mass%</th>
<th>FRAM Uncertainty</th>
<th>Run-to-run Variance</th>
<th>$^{240}$Pu Mass%</th>
<th>FRAM Uncertainty</th>
<th>Run-to-run RSD</th>
</tr>
</thead>
<tbody>
<tr>
<td>1000</td>
<td>92.87</td>
<td>2.27%</td>
<td>.84%</td>
<td>6.40</td>
<td>26.64%</td>
<td>14.10%</td>
</tr>
<tr>
<td>2000</td>
<td>94.52</td>
<td>1.44%</td>
<td>1.67%</td>
<td>5.00</td>
<td>26.36%</td>
<td>32.92%</td>
</tr>
<tr>
<td>3000</td>
<td>93.40</td>
<td>1.22%</td>
<td>1.40%</td>
<td>6.28</td>
<td>15.30%</td>
<td>18.68%</td>
</tr>
<tr>
<td>3600</td>
<td>94.35</td>
<td>1.11%</td>
<td>1.02%</td>
<td>5.35</td>
<td>18.04%</td>
<td>17.32%</td>
</tr>
<tr>
<td>4000</td>
<td>94.42</td>
<td>1.17%</td>
<td>1.02%</td>
<td>5.32</td>
<td>16.29%</td>
<td>17.70%</td>
</tr>
<tr>
<td>5000</td>
<td>93.75</td>
<td>0.98%</td>
<td>0.52%</td>
<td>5.92</td>
<td>12.74%</td>
<td>10.89%</td>
</tr>
<tr>
<td>6000</td>
<td>93.55</td>
<td>0.78%</td>
<td>0.57%</td>
<td>6.17</td>
<td>11.18%</td>
<td>8.85%</td>
</tr>
</tbody>
</table>

The Run-to-run variance is the best indication of the total uncertainty in the calculated result. However, this value is only calculable when a series of replicate runs are taken on the same item. This is not practical during an extended measurement campaign, where the intent is to minimize the time spent measuring each individual item.

Figure 5 compares the FRAM uncertainty values to the run-to-run RSDs. The error bars for the FRAM uncertainty values were calculated by taking the standard deviation of the uncertainties calculated by the software at each run time and dividing by 5 (the number of runs minus one). The error bars for the run-to-run RSDs were calculated using the formula,$^{4}$ $\text{Var}(S^2) = 2\sigma^2/(n-1)$. The standard deviation of the $^{240}$Pu mass percent values was the estimator for $\sigma$. Since $\sigma$ was estimated by the standard deviation in mass percent values, and not the relative standard deviation, the $\text{Var}(S^2)$ was divided by the average mass percent value to convert $\text{Var}(S^2)$ to a relative error. The relative error values are plotted as the one-sigma error bars in Fig. 5.

![Fig. 5. Precision.](image)
It is apparent that the FRAM-calculated uncertainty is a valid approximation of the run-to-run RSD for run times greater than 2000 seconds. These results also indicate that run times greater than 6000 seconds should be used to be confident of meeting the 10% uncertainty goal for this source/detector configuration.

As shown in Fig. 6, all of the average $^{240}$Pu values are within one RSD of the nominal value. The best $^{240}$Pu precision obtained in the experiment was 8.9% (run-to-run variance), and required a 1.7-hour count. Items with greater than 1.5 kg mass will increase the count rate, but the effect will be less than a linear function of the mass, because of self-attenuation and scattering within the packaging. The precision of the measurement for a given item can only be improved by obtaining more counts in the 642.48 keV peak, either by modifying the container, using longer count times, or using a more efficient detector. Modifying the container is currently not a realistic solution. Longer count times are possible, but undesirable and inefficient. The best solution would be the use of a more efficient detector.

CONCLUSION
Accurate plutonium isotopic results can be obtained through shielding equivalent to that provided by highly attenuating shipping containers. Attainable precision depends on the time available for the measurement. Approximately 2-hour measurements are necessary to obtain $^{240}$Pu precision less than 10% for a 10% efficient detector and a 1.5 kg item. This is due to the presence of only one weak $^{240}$Pu peak for analysis. Obtaining less than 10% precision for larger items should require less time. The precision of measurements on items of all masses would be better with a higher efficiency detector. The precision attainable in a set time should be proportional to the amount of gamma radiation escaping the container and the efficiency of the detector. The detector used in this experiment was only 10% efficient. This detector was chosen to replicate those intended for the application investigated in this experiment. Investment in a more efficient detector will pay for itself in time saved, when actual measurements are performed.
ACKNOWLEDGMENT
This work was supported by NNSA, Office of International Safeguards.

REFERENCES

