

## Measurements conducted on an unknown object labeled “Pu-239”

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### Abstract

*Measurements were carried out on 12 November 2013 to determine whether Pu-239 was present on an object discovered in a plastic bag with label “Pu-239 6 uCi.” Following initial survey measurements to verify that the object was not leaking or contaminated, spectra were collected with a High Purity Germanium (HPGe) detector with object positioned in two different configurations. Analysis of the spectra did not yield any direct evidence of Pu-239. From the measured spectra, minimum detectable activity (MDA) was determined to be approximately 2 uCi for the gamma-ray measurements. Although there was no direct evidence of Pu-239, a peak at 60 keV characteristic of Am-241 decay was observed. Since it is very likely that Am-241 would be present in aged plutonium samples, this was interpreted as indirect evidence for the presence of plutonium on the object. Analysis of this peak led to an estimated Pu-239 activity of 0.02–0.04 uCi, or  $< 1 \times 10^{-6}$  grams.*

### Introduction

The authors were requested to conduct verification measurements on an unknown object contained within a plastic bag. Inside the bag were a metal object and a hand-written label indicating “Pu-239 6 uCi,” as depicted in *Figure 1*. The object had been discovered in a storage area with several other small sources. Verification measurements were carried out on the other source materials as well.

### Measurements

The sample measurements were carried out in a conference room, where all of the sources were brought in, surveyed for contamination, and measured with two High Purity Germanium detectors (HPGe). In addition to this, the unknown object was subjected to some follow-on measurements under carefully controlled conditions.

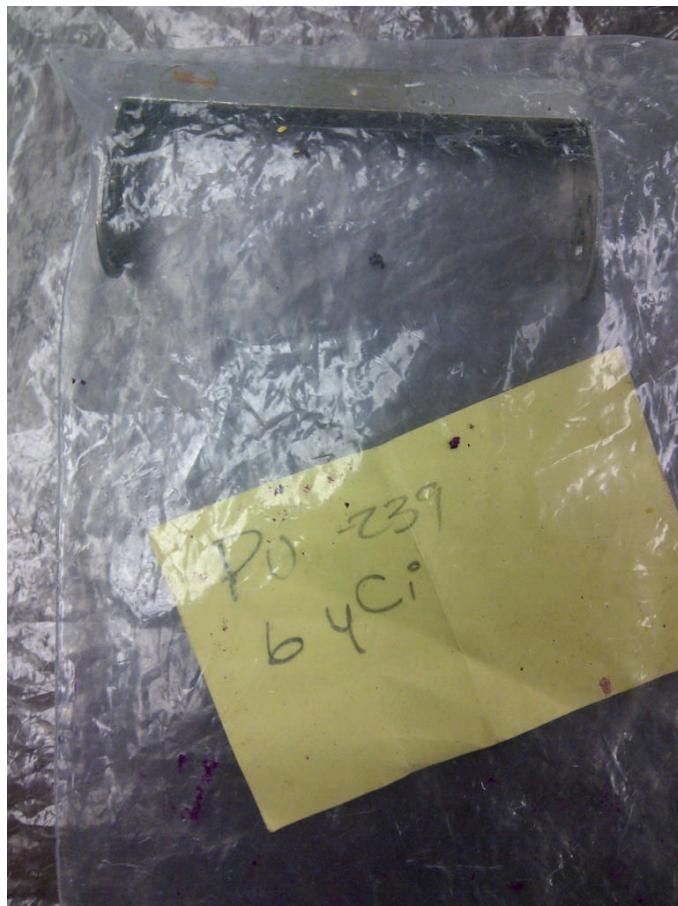
#### *Sample swipe measurements*

Since the source materials were all of uncertain origin and had been stored together under uncertain conditions, initial measurements were carried out to ensure that contamination was absent on each sample. To accomplish this, swipe samples were

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collected from the outer containment bags for each source and monitored for alpha and beta activity with the use of an ADM-300 with alpha and beta probe attachments. The results indicated that none of the source materials was leaking or contaminated.

Since the unknown object was contained within two bags, once it was verified that the outer bag was not contaminated, the inner bag was removed and surveyed with additional swipe measurements, and the object was monitored directly for beta activity while contained within the inner bag. When these measurements also did not give any indication for activity, a swipe was collected from the interior of the inner bag, which also returned null results for alpha and beta probe measurements.



*Figure 1: Picture showing the unknown object with hand written label indicating the possible presence of Pu-239.*

*Initial gamma survey measurements*

Initial gamma survey measurements were collected with a 95% liquid-cooled HPGe and a mechanically cooled Detective EX-100 positioned side-by-side for redundancy. Results from these measurements are summarized in *Table 1*.

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*Table 1: Summary of HPGe measurements verifying presence of isotope labels on the unknown sources.*

Sample Bag #	Isotope Label	Spectral Verification
1	Ra-226	Y
2	Ra-226	Y
3	Eu-152	Y
4	Th-232	Y
5	Pu-239	See discussion
6	Uranyl Nitrate	Y (U-238)
7	Depleted Uranium	Y (U-238)

### *Unknown object measurements*

To facilitate a quantitative interpretation of the measured data, follow-up measurements were carried out on the unknown object with a more well-defined source-detector geometry. These measurements were carried out with the 95% liquid-cooled HPGe detector with 7.93-cm diameter.

Two separate measurements were conducted on the object, representing two different object configurations. In the first configuration, an orange mark on the object was positioned facing the detector, whereas in the second configuration this mark was facing away from the detector. For both measurements, the object was positioned against the foam covering around the face of the detector, so that the source-detector distance was approximately 3 cm.

Each measurement was carried out for 900 seconds, real time. After the measurements were carried out, background data were collected at the same detector position and with all sources removed from the room. Unfortunately, during the configuration #1 measurements, there were a few sources present nearby, which were removed prior to the configuration #2 measurements. Thus, several peaks from Ra-226 are prominent in the net spectrum from configuration #1.

### *Detector efficiency measurements*

Additional measurements were carried out on Pu-239 and Am-241 sources with known activity in the laboratory in order to characterize the intrinsic detector efficiency for several of the gamma-ray energies associated with the decay of these radioisotopes.

## **Analysis of Gamma-ray data**

To evaluate the presence of Pu-239 on the unknown object, seven regions of interest (ROIs) were defined around peak energies associated with Pu-239 decay. In addition, an ROI was set up around the 60-keV peak associated with Am-241 decay.

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Analysis of the configuration 1 data was complicated by the presence of source materials in the background, which were not present during the background measurements. Nevertheless, it is clear from the spectra that peaks are absent within all of the ROIs. The MDA at 95% confidence for this measurement was determined through propagation of the error determined in the source and background spectra, using the relation,

$$MDA = 1.645\sigma_{net}$$

$$\sigma_{net} = \sqrt{\sigma_{src}^2 + \sigma_{bg}^2}$$

The MDA values computed for several Pu-239 photo peaks are listed in *Table 2*.

*Table 2: Minimum detectable activity determined for gamma rays associated with Pu-239 decay.*

Peak Energy (keV)	Config. 1 MDA (uCi)	Config. 2 MDA (uCi)
98.8	0.4	0.2
129.3	1.2	0.8
203.6	6.1	3.8
332.8	5.5	2.8
345.0	5.7	2.8
375.1	2.2	1.3
413.7	2.1	1.0

The smallest MDA listed in *Table 2* is from the 98.8-keV gamma ray. However, this value is determined with the assumption that the peak does not show significant overlap with other photo peaks that may be present in or around the ROI. In fact, the region around this photo peak is somewhat complex with respect to the multitude of gamma-ray and x-ray peaks that may be present here. In the configuration 2 measurements, this ROI was the only one to show greater than zero net counts under the 95% confidence criteria, with  $292 \pm 72$  net counts. However, it is questionable whether this can be interpreted as evidence for a Pu-239 photo peak given the complexity of this spectral region for most source materials.

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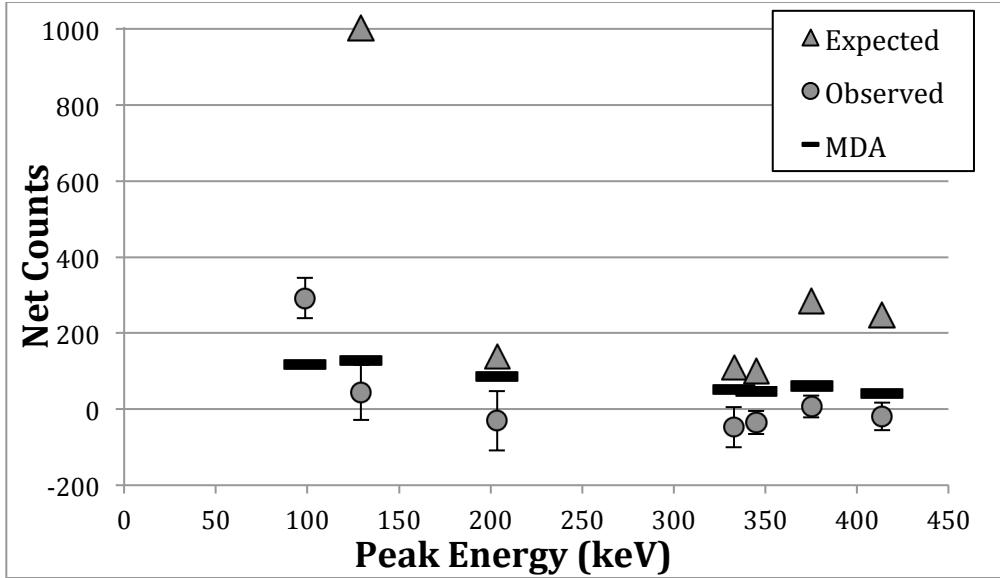


Figure 2: Plot summarizing the MDA, expected activity given a 6 uCi source and the net counts measured at peak energies associated with Pu-239 decay.

To address this issue further one can predict, given the source activity declared on the object, the measurement geometry, and the intrinsic detector efficiency measured in the laboratory, the number of counts that should be present in each photo peak. In the case of the 98.8-keV line, this calculation leads to a prediction of 2986 counts in this photo peak, an order of magnitude larger than what was observed in the spectrum.

A summary of the analysis for configuration 2 measurements is depicted in *Figure 2*. Here, the MDA determined from the spectrum is presented as black bars, the activity expected from a 6 uCi source is represented by triangles, and the measured activity for each photo peak is given as circles. As noted above, only the 98.8 keV photo peak deviates from zero to a degree that can be considered statistically significant. However, given that this spectral region can be prone to a multitude of overlapping peaks, and that no other peak shows evidence for the presence of Pu-239, this is not considered to be direct evidence for the presence of Pu-239.

#### Am-241 peak analysis

Whereas there was no direct evidence for Pu-239 present in either spectrum, the configuration 2 measurements did give some indication for a peak at 60 keV. This peak energy is associated with Am-241, which is typically present in aged plutonium samples, as it grows in from decay of Pu-241. Since the amount of the Am-241 present in a plutonium sample depends on the quantity of Pu-241 in the original sample as well as the time since separation, it is difficult to make a precise quantitative extrapolation to Pu-239 quantity from this activity. This being said, an approximate activity can be derived if one considers the potential variability in this value given the conditions noted above. *Figure 3* shows the derived plutonium

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activity from the peak area measured in the configuration-2 spectrum based on three hypothetical plutonium isotopic distributions<sup>1</sup>. The examples range in Pu-239 concentration from 93% down to 68%. Assuming that the sample was separated approximately 20 years ago, the derived Pu-239 activity ranges from 0.02 to 0.06 uCi. If one assumes a separation time greater than 30 years ago, the range converges to 0.02 to 0.04 uCi. Given a specific activity for Pu-239 of 62000 uCi/g, this corresponds to approximately  $3 \times 10^{-7}$  g of Pu-239.

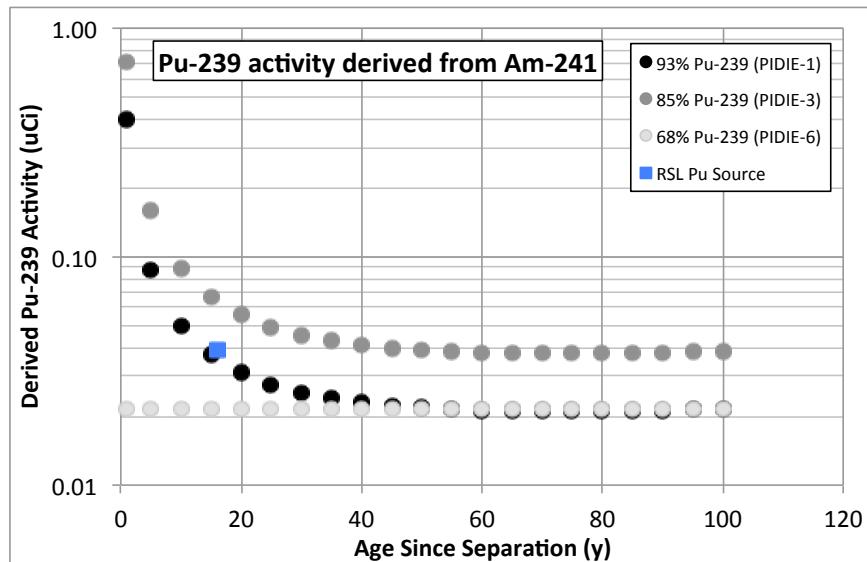


Figure 3: Estimated activity of Pu-239 derived from peak area at 60 keV for plutonium sources with different isotopic distributions.

## Summary

Measurements were carried out on an object with a hand written label indicating the presence of 6 uCi Pu-239. High Purity Germanium measurements conducted on the sample did not appear to confirm the statement, as no direct evidence of Pu-239 was evident from the spectrum. However, a small amount of activity at 60 keV did give some indication for the presence of Am-241, which could be indicative of the presence of a small amount of Pu-239. Based on the measured activity at 60 keV, the estimated quantity of Pu-239 on the object is not more than 0.04 uCi, or  $1 \times 10^{-6}$  grams.

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<sup>1</sup> The hypothetical plutonium isotopic distributions were taken from real plutonium samples.