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# A Study on the In-growth of Uranium Daughters' Gamma-rays from a Freshly Separated Uranium Gamma-ray Standard

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## Abstract

Uranium gamma-ray isotopic analysis codes rely strongly on an assumption that the uranium daughters are in secular equilibrium with their parents. Without a prior knowledge of the separation time, it is possible that freshly separated natural uranium samples can be analyzed mistakenly as enriched uranium samples using only the gamma-ray signatures. In this study, we used a 1g 0.5% uranium standard from the National Bureau of Standards (NBS) and performed a chemical separation of uranium from their daughters. The separated uranium solution was divided equally into two samples, one counted on a coaxial HPGe (High-purity germanium) detector and the other counted on a planar HPGe detector. The gamma-ray data were collected with list-mode data acquisition systems. We have analyzed the gamma-ray data with MGA++ and observed the "time-dependent" uranium analysis results, and as expected, the results are ranging from highly enriched uranium to slightly enriched uranium. However, with a proper knowledge of the separation time, the isotopic analysis results are in good agreement with the declared values (i.e., depleted uranium). We also observed gamma-rays that were signatures from the separation chemistry.

## Introduction

Gamma-ray spectrometry has been used to for determining uranium enrichment. A widely used method for this uses the uranium enrichment meter principle. This method uses a collimated detector to determine the enrichment of "infinitely thick" samples by measuring the count rate of the 185.7-keV gamma ray from the decay of  $^{235}\text{U}$ . The technique requires standards of the various enrichments and sample types. These standards are analyzed destructively with mass spectrometry to so that appropriate calibration factors used in the enrichment meter software can be obtained. For samples that are not infinitely thick, additional information is necessary to determine the enrichment.

One can also determine the enrichment by using ratios of gamma ray intensities from  $^{235}\text{U}$  and  $^{238}\text{U}$ . Unfortunately, there are no reasonably intense gamma transitions emitted directly from  $^{238}\text{U}$ , therefore, the existing techniques use the gamma rays from decay daughters of the isotopes of interest. To obtain precise enrichment results with these techniques either the daughters need to be in secular equilibrium with parents, or previous knowledge of the time of separation of the sample is required.

Figure 1 shows an example of a truncated decay chain in U isotopes: the U isotopes decays to Pb isotopes through a series of alpha and beta decay and emit gamma-rays though the decay process, the figure only shows the uranium isotopes decay to the Th-Pa daughters.

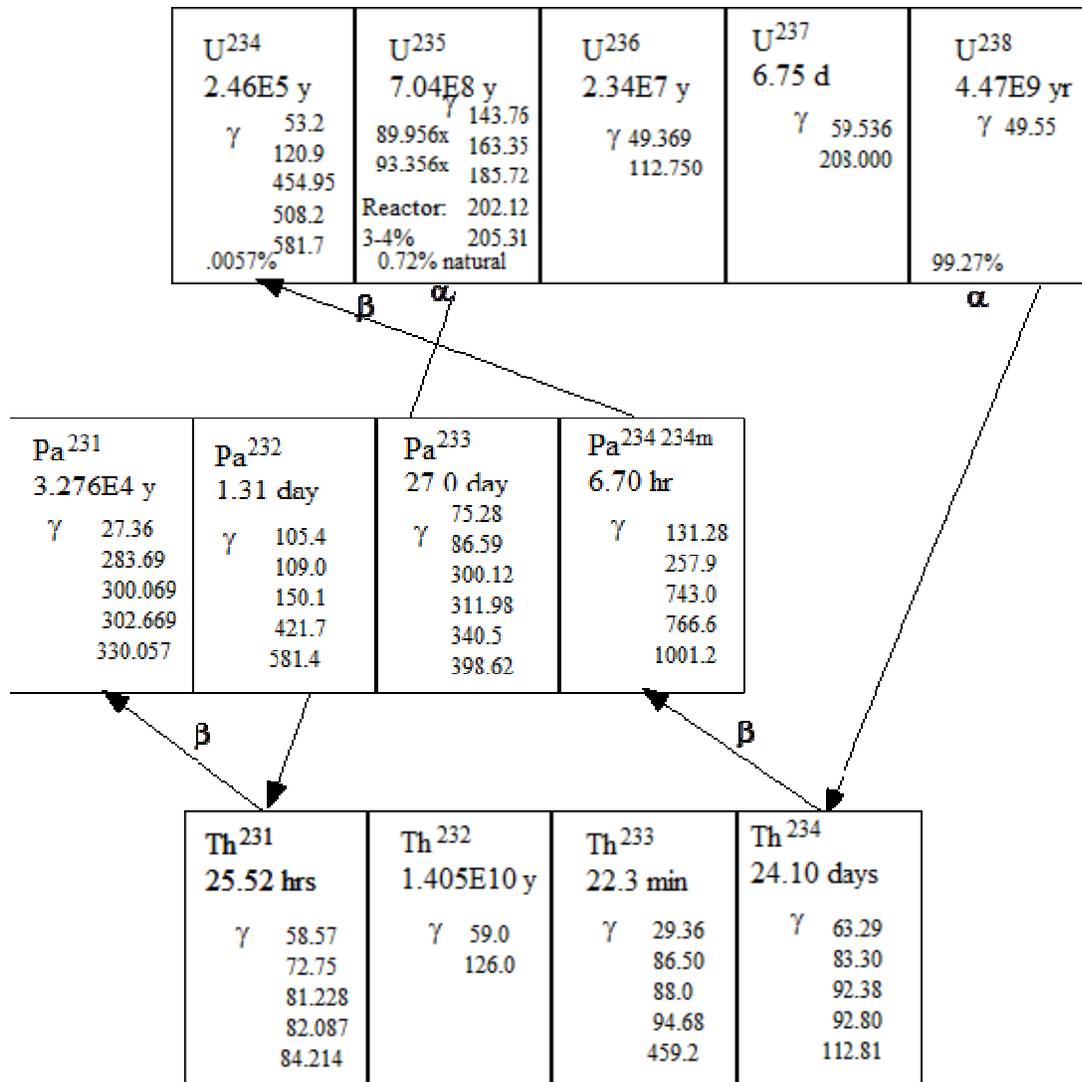


Figure 1 An illustration of truncated uranium decay chains, strongest gamma-rays (in keV) from the decay chains are also tabulated.

Commonly used isotopic analysis codes such as MGA++ (ref. 1) and FRAM (ref. 2) both use the gamma-ray intensity method; therefore, both codes could easily produce incorrect isotopic ratios if the parents and daughters were not in secular equilibrium. An example of using the Th daughter gamma-rays in the 100-keV region to determine the  $^{235}U/^{238}U$  isotopic ratio in MGA++ is illustrated in Figure 2.

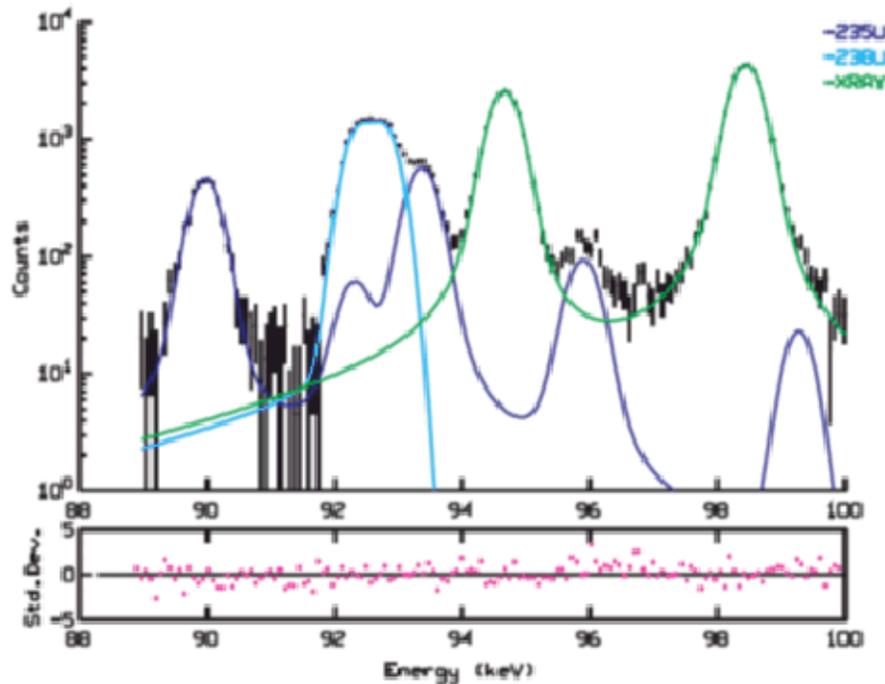


Figure 2, MGA++ unfolding of 90-100 keV gamma-ray region from uranium decay, the light blue doublet fit that representing the intensity of  $^{238}\text{U}$  was actually derived using its  $^{234}\text{Th}$  daughter gamma-rays intensities at 92.38 keV and 92.80 keV.

In this study (which is the first part of a series of studies), we will monitor the ingrowth gamma-ray intensities of the  $^{238}\text{U}$  daughters by using a slightly depleted “Standard Reference Material” SRM U-005 ( $^{235}\text{U} \sim 0.5\%$ ) gamma-ray standards from NBS (ref. 3). We will compare gamma-rays before and after uranium separation chemistry; analyzing the data with MGA++ with and without knowledge of separation time.

### Sample preparation and experimental setup

The 1g SRM-U005  $\text{U}_3\text{O}_8$  powder was dissolved in the hydrofluoric acid after ammonium-hydroxide was dissolved in hydrochloric acid. The liquid passed through the anion ion-exchange column and the uranium isotopes were separated from most of their daughters. Uranium isotopes were then extracted from column.

Uranium solution of about 20ml was divided equally into two samples, one counted on a planar detector (ORTEC GLP-36XXX) with a gain follows MGA++ specification of 0.075 keV/channel for 4k channels, the other was counted on a 30% HPGe detector (ORTEC GEM-30P4-70 ) with 0.50 keV/channel for 4k channels, NIM electronics were used for amplification and two FASTCOMTEC MPA-3 were used for list mode data acquisition. Data were collected for about two weeks and background was collected after the sample was removed for about the same length of time for background subtraction, since the  $^{232}\text{U}$  decay

chain (if there is any in the U-005 standard) was very similar to the background (mostly from the  $^{232}\text{Th}$  decay chain.)

## Data Analysis

### Coaxial HPGe:

Figure 3 shows a comparison of gamma-ray spectra of the SRM-U005 standard before and after uranium separation, as can be clearly seen that the higher energy (> 700 keV and < 1001 keV) gamma-rays (mostly from the  $^{238}\text{U}$ - $^{234,234\text{m}}\text{Pa}$ ) are missing in the fresh separated (Blue) spectrum.

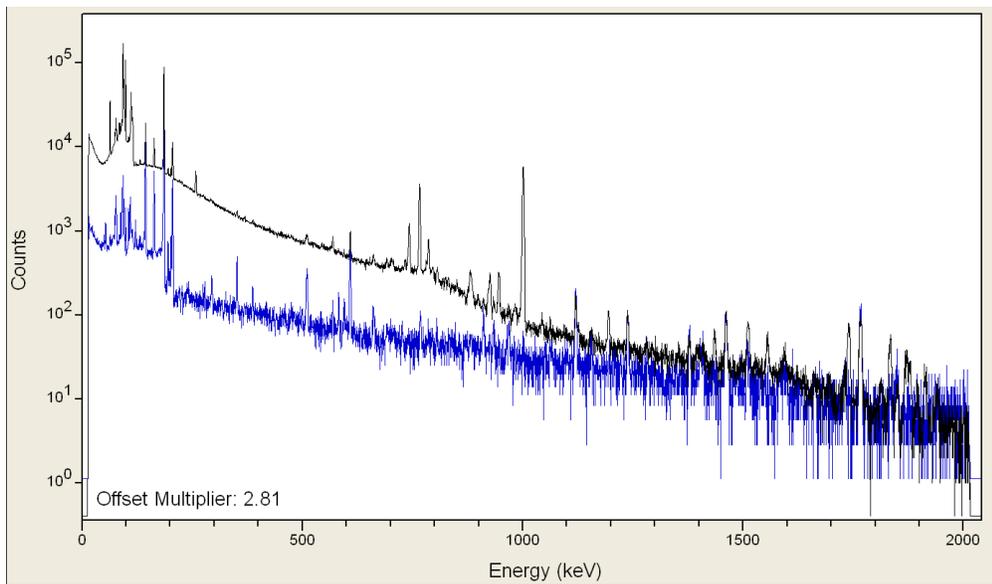


Figure 3 A comparison of SRM-U005 standard HPGe gamma-ray spectra before (Black) and after (Blue) separation chemistry.

Upon closer inspection of the lower energy (<105 keV) region, we have also observed missing (or weak) gamma ray from  $^{235}\text{U}$ - $^{231}\text{Pa}$  daughters and  $^{238}\text{U}$ - $^{234}\text{Th}$  daughters. It is clearly shown that the ingrowth of the 63.3-keV (from the decay of  $^{234}\text{Th}$ ), and 84.2-keV (from the decay of  $^{231}\text{Th}$ ) gamma-rays. Figure 3 shows a one hour data taken immediately after separation compares to a one hour data taken 44 hours after separation, and Figure 4 shows the integrated peak area of the 63.3-keV, 84.2-keV and 1001-keV from 1 hour to 36 hr after the separation. We are currently working on algorithms to take advantage of using these different ingrowth behaviors due parent-daughter not in secular equilibrium to extract time-zero information, when the separation time is unknown. Of course, the accuracy of the time-zero information will be strongly depending on the sample intensity and where in time on the ingrowth curves the sample was measured.

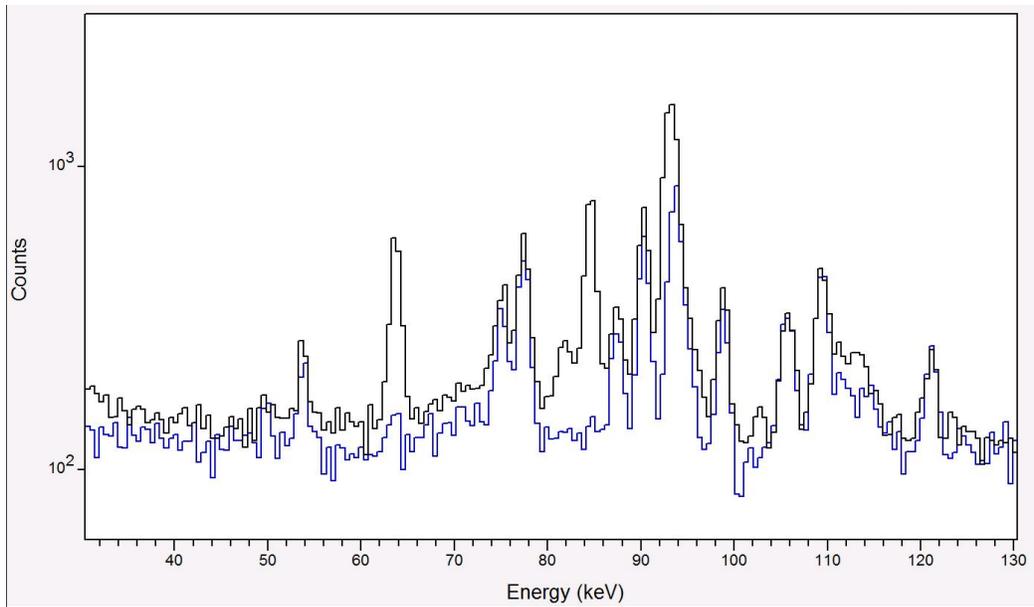


Figure 3 One hour counting after separation (Blue), one hour counting 44 hours after separation (Black), the 63-keV and the 84-keV gamma-ray intensities are increasing with respect to time.

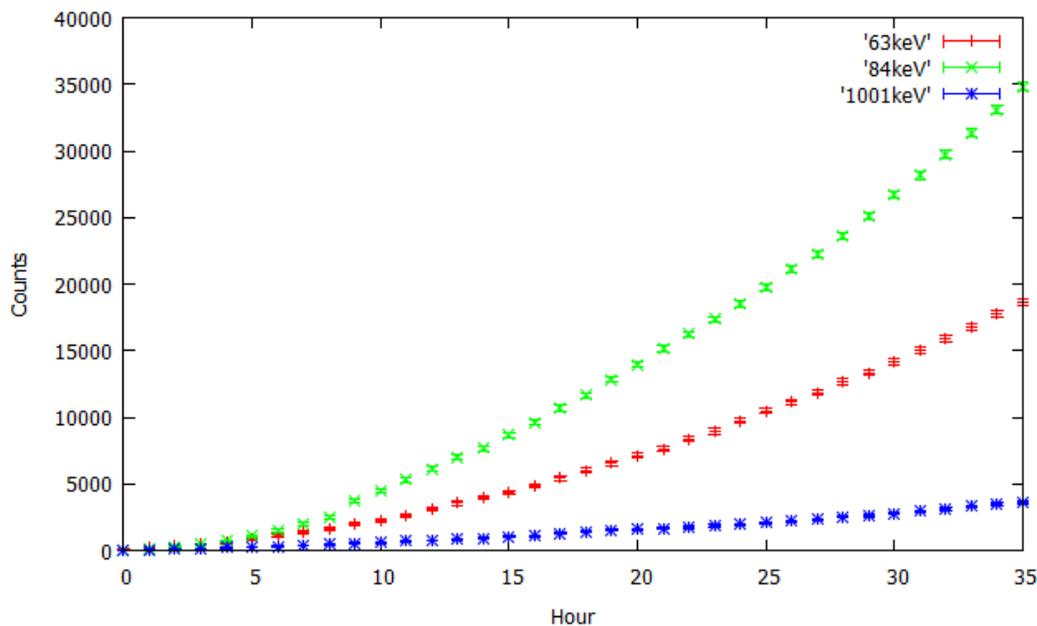


Figure 4, In-growth behaviors of the three daughter gamma-rays from the decay of  $^{235}\text{U}$  and  $^{238}\text{U}$

Planar HPGe:

The planar HPGe data were analyzed MGA++ isotopic analysis code. Because of the small sample quantity, small detector size and the MGA++ requirements of total counts in the spectrum, one day collection of data is needed. We only show couple of MGA++ analysis results, with 1-day collection time each, 1D, 5D, and 10D after time-zero, in Table 1. The first column is results based on the assumption

that parents and daughters are in secular equilibrium, the second column is applying correction to the results with knowledge of the separation time. The MGA++ produced accurate enrichment information by giving proper separation time.

| Time after separation | U-235 %   | U-235 % If the T-zero is known |
|-----------------------|-----------|--------------------------------|
| 1D                    | 23.8 +- 5 | 0.53+-2                        |
| 5D                    | 3.25 +- 1 | 0.53+-1                        |
| 10D                   | 1.87 +-1  | 0.48+-1                        |

Table 1. MGA++ analysis results

### Gamma-ray signatures that could be from the separation chemistry

To separate uranium from its daughters, depending on the quantity of the materials, different chemistry may be performed. Figure 5 shows some of the daughters maybe still associated with uranium after the process we have performed. This separation chemistry process involved hydroxide (OH-) precipitation and anion-column (Column) separation. The uranium isotopes are (Yes) for both OH- and column (i.e., stick on column). The three other species of nuclides could follow uranium isotopes are protactinium (Pa), bismuth (Bi) and lead (Pb). The isotopes of interests are  $^{234}\text{Pa}$ ,  $^{214,210}\text{Pb}$ . We have shown in the data analysis section that  $^{234}\text{Pa}$  was removed.  $^{214}\text{Pb}$  (1608s) has strong gamma-rays at 352 keV and 295 keV;  $^{210}\text{Pb}$  (20.4yr) has 49.5 keV gamma-ray;  $^{214}\text{Bi}$  (1194s) has gamma-rays at 609, 1764, 1120 keV. We have observed Pb and Bi gamma-rays in the measurements (Figure 6).

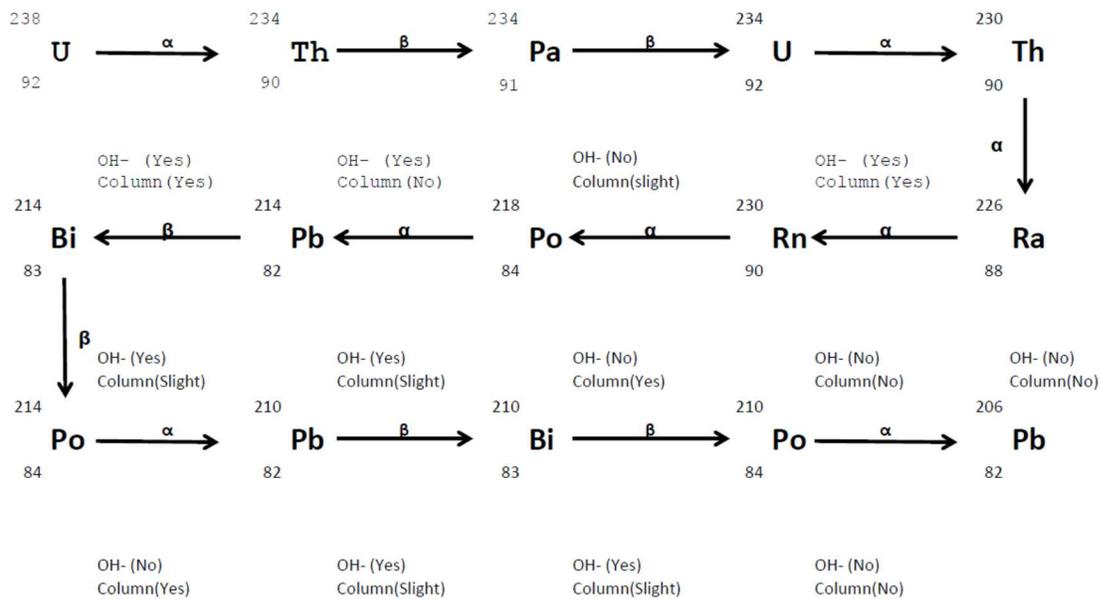


Figure 5. Possible isotopes that could follow uranium in the separation process: Pa, Pb and Po

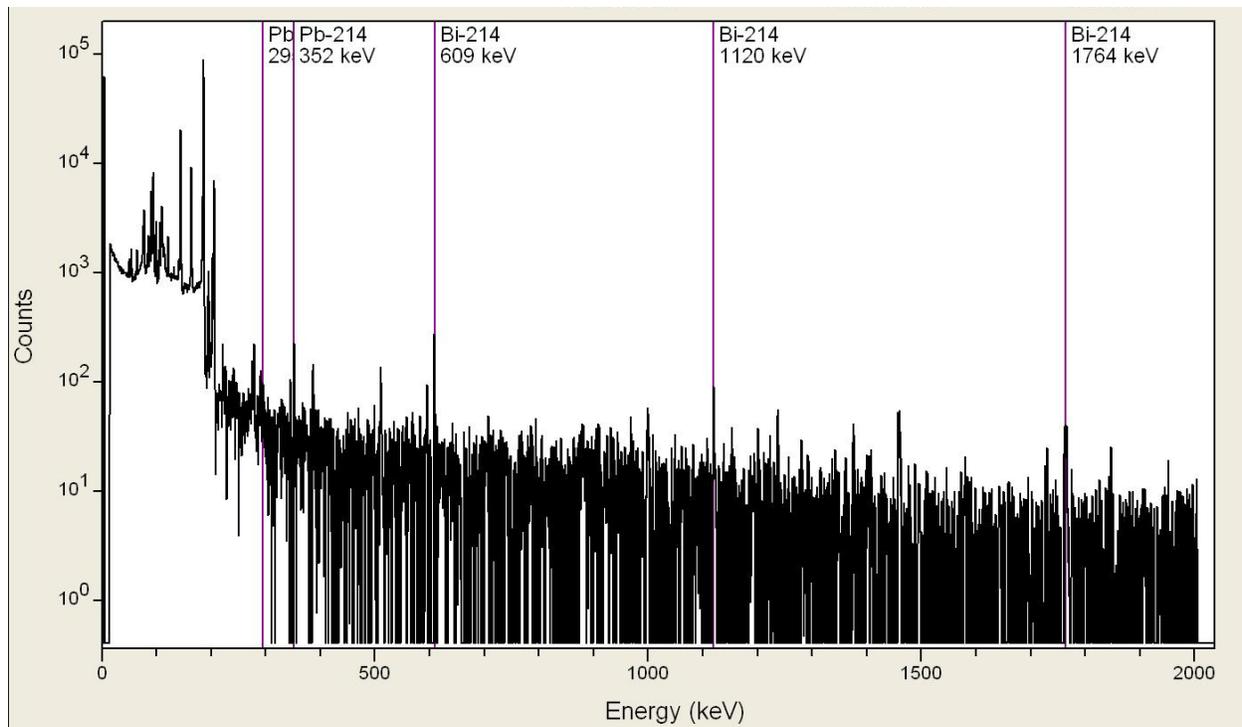


Figure 6 Background subtracted gamma-ray spectrum, the Bi-Pb gamma-rays are clearly seen

## Conclusions

With this SRM-U005 standard separation process we have shown that indeed a fresh separated depleted uranium samples can be easily misidentified as highly enriched uranium using gamma-ray spectrometry with intensity ratio method software without a prior knowledge of the separation time. We have also shown that with proper separation time, the MGA++ software will be able to obtain accurate uranium enrichment. One advantage of using list mode data acquisition is the availability of time information for each event; one can use such information and study in growth behaviors of uranium daughters to immediately determine whether the secular equilibrium condition had met. If not, from the different time characteristic of daughter gamma-ray intensities, a possible separation time can be determined and proper isotopic determination can be made. We have observed gamma-ray signatures from our chemical separation process. Bulk sample separation may show different gamma-ray signatures with different proportionality because of different separation processes were performed.

## References

- 1) "Uranium and plutonium isotopic analysis using MGA++," D. Clark, T.F. Wang, et al. , 39<sup>th</sup> Annual Institute of Nuclear Materials Management Meeting, July, 1998
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