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Abstract

The use of HPGe and gamma spectroscopy for the purpose of determining the isotopic composition of plutonium and uranium has a well-documented history in Non-Destructive Assay (NDA) and IAEA Nuclear Measurement Accountancy and Control (NMAC) measurements. Commercially available applications such as MGA/MGAU and FRAM allow for the isotopic analysis to be automated, but there are basic detector performance criteria that are typically required to produce high-quality results with low uncertainty estimates. Often the quality of an algorithm within a software application is paired with the excellent peak shape, high-efficiency, and portability of the measuring instrument. Recently, Mirion developed a new portable detector that supports both a high quality peak shape response in conjunction with relatively high efficiency. This paper presents the performance measurement results of uranium standards of varying enrichments accomplished with the Mirion Technologies Aegis[™] BE5030 portable detector and spectrometer. The isotopic and quantitative analyses were performed with FRAM. The measurements were made with Aegis[™] detectors with a range of resolution performance levels. The results of the isotopic analysis and activity complete with uncertainties are compared with those of historically acceptable laboratory instruments.

ISOTOPIC MEASUREMENTS OF SPECIAL NUCLEAR MATERIAL

INTRODUCTION

The use of High Purity Germanium (HPGe) detectors to determine isotopic composition of special nuclear material (SNM) has been well documented in Non-Destructive Assay (NDA) and IAEA Nuclear Measurement Accountancy and Control (NMAC) measurements [1]. With the development of the AegisTM portable spectrometer work to evaluate its utility to the application of isotopic analysis codes has been previously analyzed [2]. In this effort, a collection of portable HPGe detectors were used to measure uranium standards within a laboratory environment employing the use of a shield and collimator, shown below in figure 1.

A collection of 3 different Aegis units were used to make the measurements of the uranium standard. They were Broad Energy Germanium (BEGe) detectors configured with a remote detector chamber (RDC) cryostat. In each case, the units were premium resolution specifications that were all much less than 850 eV at the Co-57, 122 keV peak. Measured resolutions of the detectors are shown below in Table 1.



Figure 1: The Aegis[™] portable spectrometer, shown in the BEGe configuration with the optional RDC mounted in a shield and collimator

Resolution	Instrument
783 eV	Detector 1
823 eV	Detector 2
776 eV	Detector 3
794 eV	Average

Table 1: Detector Resolution for Isotopic Measurements

The distinct advantage of the RDC cryostat is the ability to completely enclose the detector crystal with shielding materials with a regulated entrance aperture for minimizing any environmental contamination of the measurement spectra. This is a distinction from the predecessor, the Falcon 5000[®] that could only shield along the axial direction. In this case, the measurements were made with the 50 mm lead shield, with the rear shield set to surround the detector.

Measurements were taken in 5-minute intervals up to one hour in length and the results were analyzed with the isotopic code, FRAM version 5.2 [3] or with MGAU version 4.3 [4]. With a collection of time series data, insights on the impact of adequate shielding and collimation was reduced to a measurable difference in bias and uncertainties for the measurements with no shielding or collimation and compared to the results with a completely shielded HPGe detector. For illustration the collimator was removed to show the side shielding of the RDC and is shown below in Figure 2.



Figure 2: The Aegis[™] RDC installed in the ISOCS[™] Shield with the collimator removed.

Each detector system was energy calibrated to 0.075 keV/channel, making use of 16k input channels. Each source was placed in front of the detector such that the dead time was held below 6%. Counting commenced for an hour on each sample, but the data was saved at 5 minute intervals. Measurements of uranium samples with U-235 enrichments of 0.7%, 2.9%, 4.5% and 98% were completed. Each spectrum was analyzed using FRAM version 5.2 or MGAU version 4.3.

SHIELDING AND ITS MEASURED IMPACT ON BACKGROUND SPECTRA

The most obvious and simple comparison of the shielded and bare detector measurements is to review the impact in the background that the lead shielding can have. Quantifying the change in the background, one can integrate all the counts from 50 keV to the end of the spectrum which was 1235 keV. The comparison of the large ROI, between the shielded Aegis and a bare detector, showed that the environmental background was reduced by nearly 80%, dropping the count rate from nearly 170 cps, down to about 35 cps. The visual comparison of the two spectra can be seen below in Figure 3.



Figure 3: Comparison of the shielded and bare background spectra in laboratory environment

LOW ENRICHED URANIUM MEASUREMENT ANALYSIS

The available uranium standards in the factory are limited to a small set of samples of low enriched uranium. They are uranium standards from the U.S. National Bureau of Standards. They were measured with declared enrichments of 0.7119%, 2.9492% and 4.4623% and shown below in Figure 4.



Figure 4: Uranium standards used in these measurements

Each standard was measured with the three detectors detailed in Table 1, and the results of the FRAM and MGAU analysis were normalized to the declared enrichment and averaged over all the detectors to give a single pair of data comparing the isotopic analysis of each spectra, as the count time increased.



Figure 5: Natural uranium sample measured with and without shielding and collimation analyzed with FRAM and MGAU

From the aggregation of the results, shown in figure 5 above, there is interesting divergence in the results reported by FRAM and MGAU. Both FRAM and MGAU show that the measurements taken outside the shield and collimator seem to converge to a biased result, whereas the measurements taken with the RDC enclosed in a shielded collimator have a much smaller bais. The average bias of the shielded and unshielded measurements is evaluated to be 0.1% and 5.5% respectively with FRAM and for MGAU it was 2.4% and 5.3% respectively. As to the uncertainty, the difference was less distinct and the measurement time was the largest impact in reducing the uncertainty. One key difference was the evaluation of uncertainty in MGAU was much higher than the evaluation of uncertainty in FRAM and on average, that difference was a factor of two.

As the enrichment of the sample is increased, to about 3%, the results again show a clear indication of the improvement the shielding and collimation makes on the isotopic analysis results. An illustration of the results can be seen in figure 4 below.



Figure 6: Low enriched uranium sample at 3% U-235 measured with and without shielding and collimation

Interestingly, both shielded and unshielded measurements allow the isotopic analysis to converge within about 15 minutes. Similar to the natural uranium sample, the bias of the unshielded measurements is approximately 4% whereas the shielded measurements converge neatly on the declared enrichment. The pattern of underestimated uncertainties in the unshielded measurements continues at the higher enrichment. MGAU in this case converged in both measurement arrangements, but the convergence was much quicker with the shielded measurements.

The final low enriched uranium standard is approximately 4.5% U-235. The measurement, consistent with the other LEU samples shows a clear and immediate, advantage to shielding the detector chamber, when using either code to perform the isotopic analysis. Seen in figure 5 below, the unshielded measurement has a significant bias in the isotopic enrichment. Concurrently, the shielded measurements quickly converge on to the declared value of the enrichment.



Figure 7: Low enriched uranium sample at 4.5% U-235 measured with and without shielding and collimation analyzed by FRAM and MGAU

HIGH ENRICHED URANIUM MEASUREMENT ANALYSIS

With only one highly enriched uranium sample available at the facility, there is only one look at the performance of the system with high U-235 content. The measurements, while performed in the same methodology as the previous LEU samples, yielded a unique outcome. The results are contained in the figure 6 below.

Both measurement configurations were able to achieve the declared enrichment within the quoted uncertainty, and the average biases are quite close. The biases were much less pronounced for this sample as was the increase in the uncertainty.



Figure 8: High enriched uranium sample at 95% U-235 measured with and without shielding and collimation

MEASURING URANIUM WITH AN INTERFERING BACKGROUND

In some cases, measurements are required in situations that are further from ideal than is desired. In the laboratory, this situation was simulated by placing a higher activity Cs-137 source in the vicinity of the detector making measurements of the uranium standard samples. While there was very little direct interferences in the spectra, the impact on the measurements was very clear, and the shielded collimator had the biggest opportunity to impact the isotopic analysis.

For the natural uranium sample, both isotopic codes show a significantly lower bias in the U-235 enrichment values and a slightly lower uncertainty in the results. There was a significant difference in the ability of MGAU to converge on the declared enrichment, vs FRAM, which quickly converged to the declared value well within the determined uncertainty of the evaluation. Charts illustrating the results are shown below in Figure 9.



Figure 9: Low enriched uranium sample at natural enrichment U-235, measured with and without shielding and collimation

A similar result is seen when analyzing the 4.5% LEU sample. Where FRAM results have a quick convergence to a result that has a low bias, with the shielded measurements showing no significant bias. MGAU analysis struggles still with the contaminated background, but the measurements performed in the shielded collimator have an average bias that is half the measurements taken with the bare detector. The results can be seen in figure 10 below.



Figure 10: Low enriched uranium sample at 4.5 enrichment U-235, measured with and without shielding and collimation

CONCLUSION

After an extensive set of measurements with uranium standards using the Aegis^M portable spectrometer, and an ISOXCART 50 mm lead shield and collimator, the impact on the results of the isotopic analysis of the spectrum was quantified. In general, there was a consistent pattern when comparing the FRAM results to MGAU. Both sets of results showed that the spectra where the detector was employing the collimator and shield had a significantly lower bias. For MGAU, the count time had a small impact on the bias convergence, but a distinct improvement to the uncertainty. Whereas using FRAM to analyze the isotopic concentration showed that both the bias and uncertainty

were improved with longer counting times. Depending upon the desired accuracy, most low enrichments could achieve results for the isotopic enrichment that agreed with the declared enrichment, within the uncertainty of the evaluation, within a 15-minute measurement. Future work on the matter will focus on the latest version of FRAM and the optimizations that are possible for these samples.

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