Recommendations for determining uranium isotopic composition by MGAU

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Abstract. The study deals with testing of versions 4.0 and 4.2 of the Multi-Group Analysis for Uranium (MGAU) software. MGAU is used for determining uranium isotopic composition by means of gamma spectrometry. The aim of the study was to determine the optimal measurement conditions needed to get the MGAU results as accurate as possible. The optimal number of total counts and the optimal count rate were determined. The study also shows how the accuracy of MGAU depends on the $^{235}$U-enrichment for various total numbers of counts. The testing procedure is based on using simulated spectra generated from real spectra of certified reference materials and well characterized fuel pellets. The simulated spectra are generated by randomly sampling data from real ones by Cambio software. This approach allows producing a large number of spectra having different number of total counts to obtain statistically relevant data. More than 7000 spectra have been used in the study. The results of this work help to appropriately set up a gamma-spectrometric measurement of the uranium isotopic composition.

1 Introduction

Multi Group Analysis for Uranium (MGAU) finds application mainly in nuclear safeguards, waste characterization and combating illicit trafficking. [1] This software is routinely used by analysts and nuclear inspectors for determining uranium isotopic composition. It is of interest for them to know about optimal measurement conditions and settings to be implemented while using MGAU. The purpose of this study is to provide justified recommendations for choosing such conditions and settings so the reported results can be as good as reasonably achievable.

MGAU analyses gamma ray spectra of uranium and as a result provides its isotopic composition. It can determine abundances of $^{235}$U, $^{238}$U and $^{234}$U. In some cases MGAU can estimate the abundance of $^{236}$U and detect the presence of $^{232}$U. The code allows analysing samples from depleted to 93\% enriched uranium. [2] The use of the software is very simple and straightforward. While other techniques require calibration, MGAU doesn’t need calibration prior measuring in the standard mode. Since it is fully non-destructive method, there is no sample preparation needed. [3]

There are some requirements for using MGAU. The software requires an HPGe (High Purity Germanium) detector with resolution below 750 eV at 122 keV. Ideal detector gain should be set at 0.075 keV / channel. No plutonium or other interfering isotopes can be present in a sample to be analysed by MGAU. As the software uses low energy gamma and x-ray peaks, the sample container thickness is typically limited to 10 mm of equivalent steel. [3] These basic requirements should be always applied for MGAU to work properly.

There are other settings and conditions which influence the accuracy of the MGAU results. In this work, the particular interest is given to the optimal number of total counts and the optimal count rate. The study also deals with comparing MGAU versions 4.0 and 4.2. The results of the work may serve as guidelines for analysts to determine uranium isotopic composition by MGAU. At JRC-ITU the software is used as accredited method and the results of help to update internal working instructions.
2 Material and equipment

In the study, the standalone versions of MGAU 4.0 and 4.2 have been used. A planar HPGe detector (Canberra GL0210R, S=200mm², d=10mm, Al window) has been employed for spectra acquisition. The detector was connected to Canberra Lynx electronics. The electronics were set to 5.6 microseconds rise time and 1.2 microseconds flat top. With this settings the resolution (FWHM) was 525±3 eV at 122 keV. The detector crystal and samples were surrounded by 5 cm thick lead bricks. Additional 2 mm thick Cu sheets were used to filter lead x-rays. The 185.72 keV peak of $^{235}$U was checked to be at channel 2476 corresponding to the required detector energy calibration gain 0.075 keV/channel.

Following samples have been used in the study:

- EC Certified Nuclear Reference Material 171 (U$_3$O$_8$ powders) with various enrichments (from 0.7% to 4.5%) [4]
- 6 NBS standards (U$_3$O$_8$ powders) having various enrichments (from 3% up to 90% ) [5]
- CRM 125A uranium certified fuel pellet [6]
- Two in-house characterized LEU fuel pellets [7]

3 Optimal number of counts

Gamma ray spectra are composed of several statistical events (gamma and x-ray peaks/count). This is often expressed as counting statistics. The longer a gamma spectrum is measured, the more counts are recorded. Having more counts in a spectrum means better counting statistics. This is dependent upon time for which the spectrum has been recorded. Hence, the longer a gamma spectrum is recorded, the more information is collected and the better the analysis results should be. However, due to the uncertainty of the analytical method itself, after a certain acquisition time, the results do not improve any more or do not improve reasonably. The target of this study was to find such "an optimal acquisition time" after which the MGAU results become steady.

The acquisition time is better to express in total number of counts due to different factors affecting the total time of acquisition (e.g. sample activity, measurement geometry, shielding, detector efficiency etc.). Therefore, a term "optimal number of counts" is used in this study. For complex and exhaustive testing one needs to have a large amount of gamma spectra covering wide range of $^{235}$U enrichments. Covering not only different kinds of samples (from depleted to highly enriched uranium) but also different numbers of total counts for each of the samples would be almost impossible with real measurements. Hence, the following approach has been applied.

So called “daughter spectra” have been used in the study. They were generated from real spectra recorded in a laboratory. The real "mother spectra" were recorded for very long period of time (for about 1 week). They were then used for generating the daughter spectra having different number of total counts. This way one can obtain enough spectra for providing "statistically strong" results. The advantage of these "daughter" or "simulated" spectra over spectra generated by e.g. the MCNP code is that they are based on real ones and so they contain real bits of information (i.e. laboratory conditions, background interference, electronic interference and etc.).

3.1 Procedure for determining the optimal number of total counts

Figure 1 illustrates the testing procedure. Spectra of the certified reference materials and fuel pellets were recorded in a laboratory by the high-resolution gamma detector. These so called mother spectra then served for generating corresponding daughter spectra having different number of total counts. This was done using Cambio software which allows generating simulated spectra from a real spectrum. [8] For each sample, 12 sets of 50 spectra were generated, having 12 different numbers of total counts. It means that for each sample, 50 spectra having 3.00E+05 total counts were generated, 50 spectra having 4.32E+07 total counts were generated and so on. Totally around 7000 simulated...
spectra were produced and subsequently analysed by MGAU. The results of the MGAU analysis were then compared with the certified abundances.

For comparing the results, so called MARD value was utilized. The abbreviation stands for Mean Average Relative Difference and is described by Equation 1. To calculate the MARD, first the relative difference between measured and declared abundance of $^{235}\text{U}$ is determined. Then, the absolute value of this difference is taken and finally a mean for N spectra is calculated. In this particular case N is equal to 50 spectra.

$$MARD = \frac{\sum_{i=0}^{N} |M_i - D|}{N}$$

Where

- $M_i$ – Measured value
- $D$ – Declared value
- $N$ – Number of spectra

Equation 1 Mean Absolute Relative Difference (MARD). An indicator which has been used for comparing the MGAU results with certified data of uranium abundances.
In order to get the optimal number of counts, it was necessary to take a look at the development of the MARD values over the whole range of total counts. Differences between consecutive steps were calculated and plotted as it is schematically described in Figure 2. When the differences between consecutive steps become very low, it means that the MGAU results (expressed by the MARD) do not change anymore. From this point onward, there is no sense to carry on recording gamma spectra because MGAU results do not improve anymore or do not improve reasonably.

3.2 Results: optimal number of total counts

Following graph (Figure 3) shows, that above 4.32E+07 total number of counts the MGAU results become nearly invariant and do not improve reasonably. Before this point, the values are spread over due to poorer counting statistics. From this point onward the differences between the consecutive steps are almost negligible. In addition to this, most of the biases (expressed by the MARD) lie below 1%.

One cannot reach significantly better results by performing longer acquisition Therefore it is recommended to acquire a gamma spectrum until reaching roughly 4.32E+07 total counts. This value is estimated to be the optimal number of total counts.

The detector properties are strictly defined by the basic requirements for the applicability of MGAU, so that the differences between various detectors used for MGAU are quite small. Therefore, it is expected that changing the detector would not affect the conclusion regarding the optimal number of total counts for MGAU. However, for any other software the conclusion might be different.

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1 This cannot be seen in the Figure 3 and due to the page limit the corresponding graph is not provided in this paper.

2 The stated optimal number of counts has its own statistical uncertainty, which is determined by the size of the step between the numbers of total counts used in this work. By taking smaller steps, in principle, the value for the optimal number of total counts could be refined.
There was always a question how far a sample should be placed from a detector window to get the results as accurate as possible. The sample-to-detector distance is directly related to detector’s count rate. The count rate, expressed as the number of counts per second (cps), has an influence on MGAU results. Changing the count rate affects the precision of MGAU analysis. The purpose of the testing was to find the optimal count rate with respect to the accuracy of MGAU results.

4.1 Procedure of determining the optimal count rate

In order to test the influence of count rates on MGAU results, an approach similar to the one in the previous chapter was employed. The certified materials CBNM 071, 194, 295 and 446 were measured for very long time (until reaching 2E+08 total counts) at various count rates. The count rates were set by adjusting the sample-to-detector distance. Corresponding simulated spectra were then generated and subsequently analysed by MGAU. The results of the analysis were compared with the certified data. However, instead of using the MARD value, a relative bias from declared values was calculated to find the optimal count rate. Because a bias is either positive or negative it provides useful information whether the software overestimates or underestimates the abundance of $^{235}$U.

4.2 Results: optimal count rate

The relative biases were plotted as a function of count rate (see Figure 4). Each point in the graph represents the average relative bias of a set of 50 spectra. Each set of the spectra has a certain number of total counts listed in the graph's legend. The points in the graph were fitted with a linear function and the intersections with zero were calculated for each of the trend lines. The intersections with zero represent the desired optimal count rate. As the count rate increases the bias lines go lower down from positive to the negative values. This trend is common for all of the samples, except for CBNM 071 where all the points and their corresponding trend lines lie below or just slightly above zero.
However, the calculations show that there is a dependence of the optimal count rate on the enrichment of $^{235}$U. This dependence is unclear and it is the subject of current study. For natural uranium sample CBNM 071 the optimal count rate is fairly low equal to 151±55. Together with increasing enrichment, the optimal count rate also gets greater up to 780±75 for CBNM 446. The Table I. summarizes calculated optimal count rates and corresponding dead times for all of the samples. Having looked at this table an analyst can appropriately set the sample-to-detector distance so the optimal conditions of the acquisition will be ensured. Determining the optimal count rates over the whole range of $^{235}$U enrichments is also the subject of current research.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Optimal Count Rate (cps)</th>
<th>Corresponding Dead Time (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CBNM 071</td>
<td>151 ±55</td>
<td>0.24 ±0.14</td>
</tr>
<tr>
<td>CBNM 194</td>
<td>400 ±174</td>
<td>0.66 ±0.27</td>
</tr>
<tr>
<td>CBNM 295</td>
<td>664 ±174</td>
<td>0.98 ±0.30</td>
</tr>
<tr>
<td>CBNM 446</td>
<td>780 ±75</td>
<td>1.22 ±0.11</td>
</tr>
</tbody>
</table>

Table I. Optimal count rates and corresponding dead times

![Figure 4](image_url) The graph displays relative biases of MGAU 4.2 results as a function of count rate for CBNM 446 when the number of total counts in the spectrum is more than 2.16E+07. Each point represents average bias of 50 spectra having a certain number of total counts. The higher the count rate, the lines drop down to negative values. The bias is the lowest if the count rate is at 800 cps. For the other samples the graphs look alike.

5 Validation of the approach

The spectra generated by Cambio are similar to their corresponding mother spectra but they have less total counts. They are created by randomly sampling data from their mother spectra. All the daughter spectra take after their parent the same information on background conditions, disturbances, interference and so on. In reality, background and interference conditions are being changed over the time. Hence, the real spectra of the same sample measured in the same laboratory can differ from each other. Therefore the results of analysis could be also different. Since all the measurements have been done in a laboratory under controlled and near optimal conditions, the real spectra shouldn’t be significantly different from each other. The approach of testing the software's performance needs to be approved and validated with real spectra. The simulated spectra were tested whether they correspond to real ones.
5.1 Procedure of validation

Two certified materials have been chosen for the validation test. One sample from the enrichment range where MGAU performs the best and another one from the high enrichment range where MGAU does not perform so well. The samples were measured for certain periods of time listed in the table below (Table II.). In order to have enough spectra and to be able to calculate an average and standard deviation, each measurement was repeated ten times. Having done acquisition of all the real spectra, corresponding simulated spectra (having the same number of total counts) were generated to be compared with the real ones.

<table>
<thead>
<tr>
<th>Number of spectra recorded</th>
<th>10</th>
<th>10</th>
<th>10</th>
<th>10</th>
<th>10</th>
<th>10</th>
<th>10</th>
<th>10</th>
</tr>
</thead>
<tbody>
<tr>
<td>Acquisition Time</td>
<td>10 min</td>
<td>20 min</td>
<td>30 min</td>
<td>1 hour</td>
<td>2 hours</td>
<td>5 hours</td>
<td>10 hours</td>
<td></td>
</tr>
</tbody>
</table>

Table II. Acquisition time and number of spectra recorded for validation test

5.2 Results of validation

Following graph (Figure 5) depicts results of the validation test for one of the samples used in this test. Towards the greater number of total counts, as the statistics improves, the two points in the Figure 5 (triangle and rectangle) lie closer to each other and also their respective error bars are getting overlapped. The error bars represent the standard deviation of the MARD. There is also an apparent decreasing trend of the values along the x-axis. Having a similar decreasing trend line and having a similar standard deviation together with being in the same range, the simulated spectra correspond to the real ones in all aspects.

Figure 5 Comparison of the MARD values of real spectra versus simulated spectra. The sample comes from the enrichment range (4.46 %) where the code performs the best. For the other sample which was used in this study, the corresponding graph looks similar.
6 MGAU V4.0 vs. MGAU v4.2

The newer version 4.2 of the code became available and for users it is worthwhile to know whether to update their current version. The purpose of the comparison is to provide users with information about differences between the two recent versions of MGAU.

6.1 Procedure of the comparison

Having reached the optimal number of total counts (4.32E+07), the results of MGAU analysis do not change further with increasing the number of total counts. As the MGAU results become stable and unaffected by counting statistics the differences between the two versions of the code can be examined. Therefore, the two versions of the code were compared using spectra having the optimal number of counts. The same procedure based on the simulated spectra has been used. For benchmarking the codes, bias of averages has been used.

6.2 Results of the comparison

The results indicate that there are minor differences between the two versions. Both versions tend to underestimate the $^{235}$U abundance. However, the study shows that the individual differences between the two versions are statistically equal, as they lie within the standard deviations.

7 Conclusion

The optimal number of counts has been estimated to be 4.32E+07 total counts recorded in a gamma spectrum. The study has shown that recording gamma spectra with more counts beyond the optimal number of total counts does not generate better results.

The optimal count rate was found to be dependent on the $^{235}$U enrichment of a measured sample. This study provides a table of optimal count rates as a function of enrichment. Analysts may use this table to adjust the sample-to-detector distance to reach the optimal count rate. These values were found to be valid for the particular HPGe spectrometer used in this study. Even though the parameters of detectors to be used for MGAU are strictly done, further tests with different spectrometers and samples are the subjects of current research.

The accuracies of the results for $^{235}$U abundance reported by MGAU v4.2 and v4.0 were compared. The comparison has been done using the established optimal number of counts. Both versions showed almost identical performance.

This study also provides an overview about the expected accuracy of MGAU results for given numbers of total counts and for given $^{235}$U enrichments. The expected accuracy is provided for various numbers of total counts, ordered from the lowest to the highest and also for different $^{235}$U enrichments. Before launching the spectra acquisition analysts can learn about the expected accuracy of the MGAU analysis. The time of the acquisition can be then adjusted to their needs. Due to space limitations the results are not reported in this paper and are available upon request.

The work deals solely with the abundance of $^{235}$U as it is the most important information when analysing uranium isotopic composition. Performance of MGAU in determining abundance of $^{234}$U is the subject of current study.
8 Acknowledgment

Special thanks go to Andrey Bosko who provided us with an extremely useful spreadsheet and helped us save a lot of time and efforts.

9 Works Cited


