COMPARISON OF THE PERFORMANCE OF DIFFERENT URANIUM ENRICHMENT ANALYSIS CODES USING A RANGE OF DETECTOR TYPES

M. J. Koskelo[1] , J. A. Chapman[1] , J. D. Marsh[2] and S. W. Stevens[2]
[1]Canberra Industries, 800 Research Parkway, Meriden, CT 06450, USA
[2]Canberra Industries, 1133-C Oak Ridge Turnpike, Suite 260,
Oak Ridge, TN

ABSTRACT

This paper describes the results of a set of measurements carried out to assess the state of the practice of relative uranium enrichment measurements by gamma ray spectrometry and to compare a range of approaches by carrying out measurements on a set of uranium samples with known enrichments. Measurements made by a team from Canberra Industries were carried out using two detector/counting chain combinations. The data were collected in such a way as to allow it to be analyzed using different uranium enrichment analysis methods. The results obtained using different detector and enrichment analysis method combinations were compared with each other and with the known values for each sample. Results of these measurements show that in controlled experimental conditions the total uncertainty of reported enrichment is below the procedural upper reporting limit of ±20%. Measurements tend to be better with a BeGe detector over a coaxial detector.

INTRODUCTION

Canberra Oak Ridge, LLC is the Nondestructive Assay/Nondestructive Examination (NDA/NDE) Services Subcontractor to Bechtel Jacobs Company , the Environmental Management and Enrichment Facilities Management and Integration Contractor at Oak Ridge. The main focus of the contract was originally for NDA waste measurements in fixed facilities such as K-1423 at the East Tennessee Technology Park (ETTP), Building 9720-32 at the Y-12 Site, and the Waste Examination Assay Facility at the Oak Ridge National Laboratory. Outside this main contract, Canberra also performs numerous field measurements of waste containers at the three Oak Ridge Reservation sites. Waste characterization is one of the vital functions performed at these sites and NDA of waste containers is the primary method used for waste characterization.

An important part of the NDA measurements is the determination of uranium enrichment in the assayed samples. Historically, the total measurement uncertainty (TMU) of +/- 20% (relative) in the presence of +/- 50% absolute uncertainty has been reported at the Oak Ridge K-25 site for ^{235}U enrichment. This estimated uncertainty bound was established in the early 1990s by Martin

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Marietta Energy System's NDA measurement organization at the K-25 Site. Interviews with a former employee revealed that it was based on data analyses from a series of NDA measurements of known uranium sources. Unfortunately, documentation of this evaluation has not been found. In the mean time, the signal processing hardware and the enrichment analysis methods have evolved since the early 1990s. Therefore, we have conducted a series of new NDA test measurements of known uranium sources using presently available hardware and software to re-establish this estimated uncertainty bound.

EXPERIMENTAL

A set of uranium sources, each with a different uranium enrichment, was measured at Canberra's Non-destructive Assay facility at the Y-12 site in Oak Ridge, Tennessee. The measurements were carried out by the team from Canberra's Oak Ridge Operations, and used two different detector/counting chain combinations as follows:

- A broad energy germanium detector (BEGe), Canberra Model BE3825 and a Canberra Inspector-2000 digital MCA
- A small conventional p-type coaxial germanium detector, Canberra Model GC6020 and a Canberra Inspector-2000 MCA.

The sources with known enrichment are shown in Table 1. These NIST certified uranium isotopic standards were chosen to best represent the spread of uranium enrichment values seen in Oak Ridge waste containers. Each known source was placed inside a 5 gallon empty container and a 10 gallon empty container as shown in Table 1 with each source centered in the container. Each measurement was performed at a 12 inch standoff from the side of the container with the detector aimed at the center point of the known source inside it. The drum is not rotated.

Table 1. Enrichments of the known test sources.

Measurement	Container Size	Reference Enrichment %
A	5-gal	1.51
B	5-gal	4.95
C	5-gal	19.81
D	5-gal	49.38
E	5-gal	93.21
F	10-gal	1.51
G	10-gal	4.95
H	10-gal	19.81
I	10-gal	49.38
J	10-gal	93.21

The BEGe detector was set up to collect 8192 channel spectra with a gain of 0.1865 keV/channel. Each spectrum was collected with a live time of 30 minutes according to the presently established procedures at the Oak Ridge site and then stored using the Canberra Genie-

2000 software for later analysis. This gain setting is used for reasons of flexibility even though it is not the most common gain setting for typical enrichment analysis programs.

The coaxial detector was set up to collect 8192 channel spectra with a gain of 0.2022 keV/channel. Each spectrum was collected with a live time of 30-60 minutes according to the presently established procedures at the Oak Ridge site and then stored for later analysis using the Canberra Genie-2000 software. Each of the collected spectra was then analyzed using four uranium enrichment methodologies:

1) ISOCS V4.0[1],

2) MGAU V3.2[2],

3) Program Isotopic V2.0.6[3]

Program Isotopic software is classified as Government Furnished Equipment (GFE) and has been in use at the East Tennessee Technology Park's K-1423 facility and for HPGe field measurements since the mid-1990s.

The ISOCS method does not calculate the uranium enrichment directly. In this particular context, the ISOCS method refers to results that were derived from the quantitative ISOCS analysis results as

$$E_{SYS} = \frac{m_{235}}{m_{235} + m_{238}}$$

where m_{235} and m_{238} are the quantitative activity concentrations calculated by ISOCS converted to grams by dividing by the appropriate specific activity for ^{235}U (185 keV) and ^{238}U (1001 keV), respectively. This is an approximation and does not account for the slight (< 1%) mass contribution from ^{234}U and ^{236}U.

Since the actual samples can vary in size, we treated the two sets of results (from the 5 gallon container size and from the 10 gallon container size) as a single set of measurements. First, we compared the results to the known enrichment values. Besides the individual % differences for each measurement, we calculated an average percent difference for each of the methods as

$$D_{Av} = \frac{1}{N} \sum_{i=1}^{N} d_i$$

where N is the number of measurements (10), and d_i are the individual % differences between the calculated enrichment and the known enrichment.

In addition to knowing the average bias, or percent difference and the known values, it is also of great interest to know whether the various enrichment analysis methods report an uncertainty estimate for their results that is appropriate for the deviation from the known value. This has been tested by applying the chi squared test to the enrichment analysis results.

If the chi-squared is outside the acceptable range, the reported uncertainties cannot be used as a reliable measure of the uncertainty budget. Therefore, we also calculated the standard deviation of the percent differences. Assuming that our measurements represent independent samples of a distribution of differences from the known values, 95% of this percent difference distribution will fall within +/- 1.96 times the standard deviation. This gives us an estimate of the uncertainty budget that should be used for each of the methods.

RESULTS

The measured enrichment results and their percent differences from the known enrichment values for each of the ten measurements with the BEGe detector are presented in Table 2 for the Program Isotopic, ISOCS and MGAU methods. The three methods are compared to each other graphically in Figure 1. At the bottom of Table 2, we have calculated three different measures of the goodness of the results. The first set of values consists of simple averages of the percent difference values for each method. The second set of values consists of the standard deviation of the percent difference values of each of the methods. The third set of values consists of the chi-squared values of each method.

As can be seen from Table 2, all methods have very small average % differences. MGAU has the smallest scatter in the deviations from the known value as indicated by the entries labelled "1.96 x std dev of % differences". All methods have a scatter below 20%.

For a series of ten measurements, the chi-squared value should follow the chi-squared distribution with ten degrees of freedom. The 95% confidence interval of such a distribution is from 3 to 20. The MGAU method and the enrichment calculations based on the ISOCS quantitative results have a chi-squared value that is well within these bounds. Thus they both report results that are in statistical control, i.e. they report uncertainties that are appropriate for the deviation of the reported value from the known value. The Program Isotopic exhibits a chi-squared value that is well above the upper limit indicating that it reports an error bar that is too small for the actual difference from the correct value.

When using a BEGe detector in the configuration used in this study, it can be concluded that all three methods produce results that are within the +/- 20% uncertainty budget that has been used at Oak Ridge. It can be argued that the when using MGAU the uncertainty budget could be reduced to as low as +/- 10%. With chi-squared values of 8 and 7, respectively, MGAU and ISOCS provide results that are in statistical control. Therefore, statistically their reported uncertainties can be used as such without the use of an estimated uncertainty budget altogether. With a chi-squared value of 37, the Program Isotopic underestimates the uncertainties. Therefore, its uncertainties cannot be used as is, but using a +/- 20% uncertainty budget is appropriate.

Table 2. Uranium Enrichment Results for the BEGe detector measurement setup from Program Isotopic, ISOCS and MGAU.

CONTAINER SIZE	**Reference Enrichment**	**Program Isotopic Enrichment**	**% Difference**	**ISOCS Enrichment**	**% Difference**	**MGAU Enrichment**	**% Difference**
5-gal	1.51	1.44	-4.8	1.53	1.0	1.51	0.1
5-gal	4.95	4.28	-13.5	4.78	-3.4	4.94	-0.3
5-gal	19.81	21.68	9.4	20.78	4.9	19.33	-2.4
5-gal	49.38	53.37	8.1	54.69	10.7	46.90	-5.0
5-gal	93.21	86.92	-6.7	78.29	-16.0	92.99	-0.2
10-gal	1.51	1.25	-17.4	1.43	-5.4	1.43	-5.6
10-gal	4.95	4.50	-9.1	5.00	1.1	4.54	-8.3
10-gal	19.81	17.41	-12.1	19.45	-1.8	20.34	2.7
10-gal	49.38	45.72	-7.4	53.18	7.7	49.39	0.0
10-gal	93.21	81.98	-12.0	87.04	-6.6	94.82	1.7
Average % difference			**-6.6**		**-0.8**		**-1.7**
1.96 x std dev of % differences			**17.7**		**15.5**		**7.0**
Chi-Squared			**37.1**		**7.1**		**8.0**

Based on these results, the ISOCS and MGAU methods can be recommended for use with BEGe detectors without reservations. The Program Isotopic can be used with a BEGe detector if a +/- 20% uncertainty budget is used.

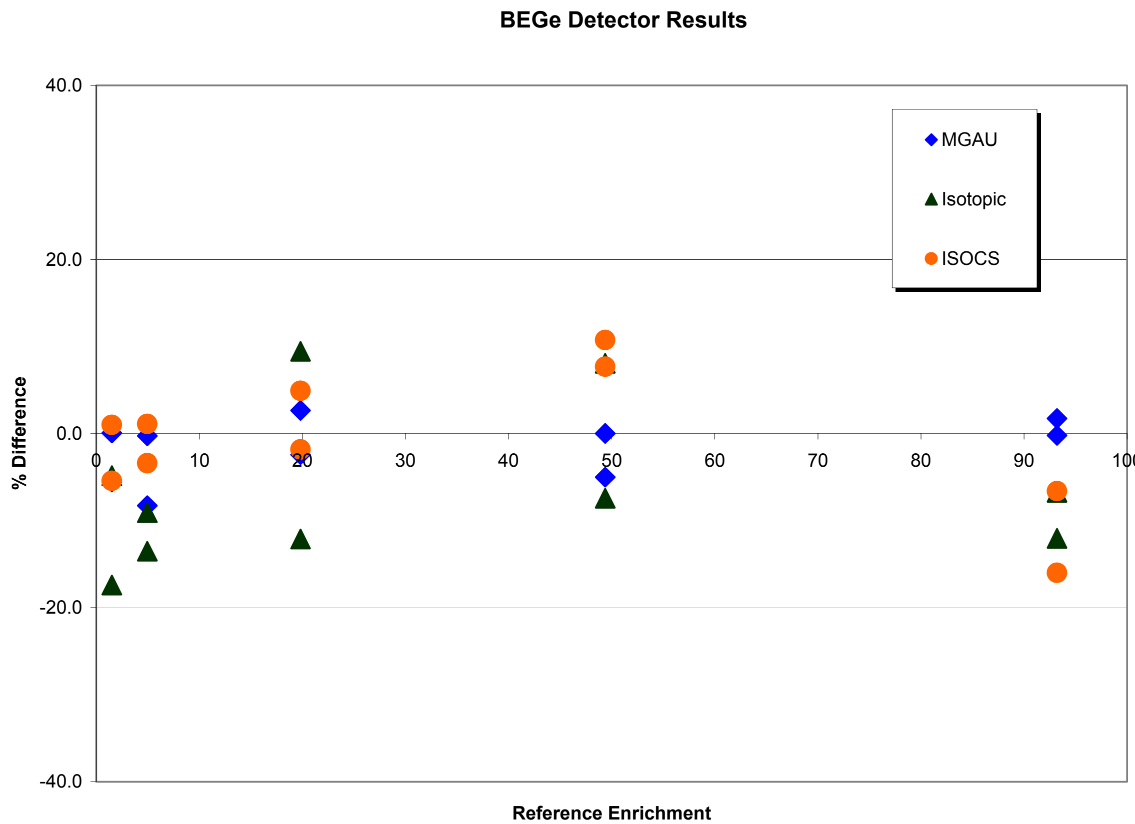

Figure 1 Comparison of enrichment results from the three methods using a BEGe detector.

The measured enrichment results and their percent differences from the known enrichment values for each of the ten measurements with the coaxial detector are presented in Table 3 for the Program Isotopic, ISOCS and MGAU. The three methods are compared to each other graphically in Figure 2. At the bottom of Table 3, we have again calculated three different measures of the goodness of the results, the averages of the percent difference values, the standard deviation of the percent difference values, and the chi-squared values of each method.

Table 3. Uranium Enrichment Results for the coaxial detector measurement setup from Program Isotopic, ISOCS and MGAU.

CONTAINER SIZE	**Reference Enrichment**	**Program Isotopic Enrichment**	**% Difference**	**ISOCS Enrichment**	**% Difference**	**MGAU Enrichment**	**% Difference**
5-gal	1.5132	1.48	-2.2	1.77	16.9	2.44	61.0
5-gal	4.949	4.71	-4.8	6.24	26.0	4.20	-15.1
5-gal	19.811	20.81	5.0	20.64	4.2	18.63	-5.9
5-gal	49.383	34.80	-29.5	44.46	-10.0	45.56	-7.8
5-gal	93.2072	92.32	-1.0	74.80	-19.7	95.29	2.2
10-gal	1.5132	1.50	-0.9	1.27	-16.2	1.05	-30.7
10-gal	4.949	4.23	-14.5	5.54	11.9	6.38	29.0
10-gal	19.811	19.30	-2.6	17.01	-14.1	17.14	-13.5
10-gal	49.383	43.05	-12.8	46.40	-6.0	45.78	-7.3
10-gal	93.2072	74.32	-20.3	76.78	-17.6	84.66	-9.2
Average % difference			**-8.4**		**-2.5**		**0.3**
1.96 x std dev of % differences			**21.3**		**32.4**		**52.3**
Chi-Squared			**6.3**		**98.2**		**42.3**

As can be seen from Table 3, MGAU has the smallest average % difference for this detector. However, the standard deviations of the % differences and the chi-squared values of the methods show a very different pattern. The Program Isotopic is the only one with an acceptable chi-squared value. Its relative scatter for the deviations from known values is just over +/- 20%. The results from the ISOCS method and from the MGAU method indicate a much larger scatter.

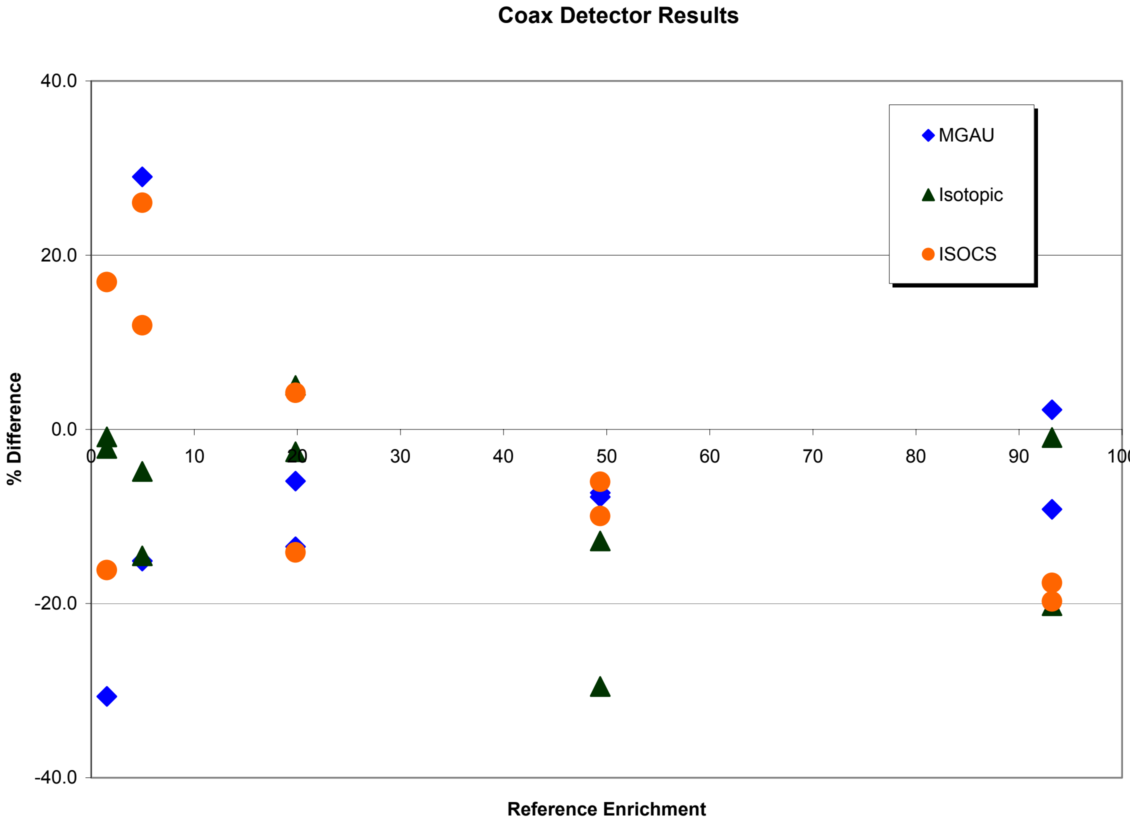

Figure 2 Comparison of enrichment results from the three methods using a coaxial detector.

Based on these results, one can conclude that the Program Isotopic can be used for enrichment measurements with coaxial detectors. Furthermore, it's results seem to support the original uncertainty budget of +/- 20% for this detector type. The ISOCS and MGAU methods are not recommended for use with a coaxial detector despite the very small average % differences.

CONCLUSION

The enrichment estimates calculated from the activity values obtained with the ISOCS calibration method indicate that this method is suitable for use with a BEGe detector. The results are in statistical control and within the +/- 20% uncertainty budget that has been in use. The ISOCS method does not appear to be suitable for use with a coaxial detector. The MGAU program provides enrichment estimates that are in statistical control when using a BEGe detector. The results are consistent with about a +/- 10% uncertainty budget. MGAU is not suitable for use with a coaxial Ge detector. This is not altogether surprising. MGAU was originally developed for low energy detectors with a much better resolution than observed for the coaxial detector used in this study. The Program Isotopic provides results for both detector types that are approximately consistent with the +/- 20% uncertainty budget that has been in use for uranium enrichment estimates on the Oak Ridge Reservation. The Program Isotopic results are in statistical control only for the coaxial detector data. In practice, it is important to underscore the procedural requirements for using a specific detector type/software combination. Additional effort is needed to assess any biases introduced in the results as a function of source configuration, source spatial distribution, waste matrix, and larger waste containers.

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