Comparison of the Performance of Different Plutonium Isotopic Codes Using a Range of Detector Types

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ABSTRACT

This paper describes the results of a set of measurements carried out as part of the ESARDA NDA Working Group's Pu-2000 Exercise. The purpose of the exercise was to assess the state of the art of relative plutonium isotopic measurements by gamma ray spectrometry and to compare a range of different approaches by carrying out measurements on a set of unknown plutonium samples. Measurements made by a team from Canberra Industries were carried out using three different detector/counting chain combinations. The data was collected in such a way as to allow it to be analyzed using a number of different plutonium isotopic analysis codes. The results obtained using different detector and isotopic code combinations were compared with each other and with the certified values for each sample.

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INTRODUCTION

Fast and accurate determination of the uranium enrichment and/or the isotopic composition of plutonium are essential in nuclear disarmament and stockpile stewardship. This includes the traditional verification and confirmatory measurements of Special Nuclear Materials (SNM) like those performed by Euratom and the IAEA, as well as Material Protection, Control and Accountability (MPC&A) applications for SNM identification at the nuclear facilities by their operators. Such measurements can generally be divided into two broad categories, low-resolution measurements and high-resolution measurements. Low-resolution measurements generally mean measurements with room temperature detectors, such as NaI or CdTe crystals. Particularly NaI systems are used extensively to verify uranium enrichments[1,2].

High-resolution systems can be used for enrichment measurements as well[3], and are routinely used for measuring plutonium isotopic compositions. Several software packages have been developed for this purpose over the years. The most well known plutonium isotopics codes are MGA[4], TRIFID[5] and FRAM[6].

The MGA code was originally developed by Ray Gunnink and co-workers at the Lawrence Livermore National Laboratory[7]. It is perhaps the most widely used code for determining plutonium isotopics. It does require a high-resolution detector, as all of the plutonium isotopics codes do. It has also been found to provide accurate results in various comparison exercises[8,9], and the results are not dependent on sample age, geometry, chemical composition or attenuation between the sample and detector.

Canberra has a license to sell and distribute MGA. It has been commercially incorporated into the U-Pu InSpector system[10]. In addition, the MGA code has been enhanced to be more robust for situations where the counting statistics are far from ideal[11]. The hardware typically used for field plutonium measurements has also been improved to provide better resolution and stability[12]. Other published improvements include a more accurate determination of the ^{237}Np content in plutonium materials[13]. Recently, a new analysis mode has been added to MGA to allow a complete analysis to be made based on only the high-energy region of a spectrum[14].

The original TRIFID package was designed by John Fleissner and his co-workers at the Mound Laboratory under the name GRPAUT[15]. The code was developed further and used extensively at the Rocky Flats National Laboratory, at which time it became known under the name TRIFID. Canberra has a limited license to sell and distribute the TRIFID code, and has an easy to use interface that ties in with the Genie-2000 software for it. TRIFID does not provide isotopic results for Uranium-only bearing materials. Its design is similar to MGA in that the decision rules on how to perform the analysis are inside the program. At the same time, it does not analyze the 100 keV region, and is thus more similar to FRAM in its analysis capabilities. Because TRIFID analyzes the high-energy gamma rays, it is inherently capable of analyzing plutonium samples with more attenuating containers.

The original FRAM software package was designed and developed by George Nelson of the University of Arizona and Tom Sampson of the Los Alamos National Laboratory. The more recent versions of FRAM have been

converted to operate on a PC under Microsoft Windows[1]. Canberra has a license to sell and distribute the Windows FRAM code. Like TRIFID, the FRAM analysis only includes peaks above the 100 keV region. Just like MGA and TRIFID, FRAM uses selected peaks in the unknown sample spectrum to provide an energy calibration, a peak resolution (FWHM) calibration, and a peak tailing calibration. Unlike MGA and TRIFID, which contains all the logic of what peaks to use and how to analyze them inside the code, FRAM has the characteristic that the peaks that are used for analysis and exactly how to analyze them are stored in tables that are outside the source code. This has the benefit that even very special situations can be accommodated, but the disadvantage that there are fewer automatic rules to direct the flow of calculations. An example set of such tables is normally distributed with the PC/FRAM package.

Even though such codes have been in existence for over ten years, there has been further development and refinement in their capabilities. In addition, there has been development in the detector technology over that period of time. Current detectors provide better resolution at the same time as they provide a higher efficiency than the detectors that were available ten years ago. At the same time, with the introduction of digital signal processing (DSP) technology into nuclear instrumentation, the performance of the electronics used in plutonium isotopics has also been enhanced. And finally, measuring plutonium isotopics has always also required some skill on the part of the operator. In order for the software to work best, the equipment needs to be set up correctly, with good resolution, peak shape, and dead time, among other things.

To establish the state-of-the-art in measuring plutonium isotopics, the ESARDA NDA Working Group organized an inter-comparison exercise, Pu-2000, carried out at IRMM in Geel, Belgium during the Summer/Autumn of 2000. A set of plutonium samples, each with a different plutonium isotopic mix were measured as complete unknowns by teams of participants from a number of laboratories worldwide, including SCK-CEN Belgium, CEA and IPSN France, Los Alamos Laboratory, Lawrence Livermore Laboratory, JRC Ispra and Canberra Industries. In this paper, we present the results from the Canberra team.

Experimental

The measurements carried out by the team from Canberra Industries used three different detector/counting chain combinations as follows:

- A low energy germanium detector (LEGe), Canberra Model GL0515R and a Canberra Inspector-2000 digital MCA.
- A broad energy germanium detector (BEGe), Canberra Model BE3820 and a Canberra DSA2000 digital MCA.
- A small (13%), conventional p-type coaxial germanium detector, Canberra Model GC1318 and a Canberra Inspector MCA.

A set of known encapsulated plutonium oxide standards, identified as CBNM61, CBNM70, CBNM84 and CBNM93, was used for setting up the equipment. A set of encapsulated plutonium oxide and/or MOX samples, each identified by letters A through T were then measured as unknowns as part of the exercise. The sequence of measurements and data analysis carried out by the various participants is as follows:

1. Participant measures all samples as complete un knowns.
2. First submission of results, which are influenced by the equipment, the evaluation method and the ^{242}Pu algorithm.
3. ^{242}Pu values are revealed by IRMM.
4. Participant recalculates results with "declared" ^{242}Pu values.
5. Second submission of results, which are independent of the ^{242}Pu algorithm.
6. IRMM reveals certified and characterized values to participant.
7. Results are submitted to CEA DAMRI by IRMM.
8. Evaluation.
9. Two sets of evaluations will be performed, with and without influence of the ^{242}Pu algorithm.

The LEGe detector system was set-up to collect 4096 channel spectra with a gain optimized for low energy MGA analysis (0.075 keV/channel). The results from the LEGe detector using the low energy MGA mode was used as the primary set of results submitted by Canberra to the Pu-2000 Exercise organizers. The coaxial detector

counting system was set to collect 8192 channel spectra at a gain oF 0.125 keV/channels, allowing it to be used by the high energy analysis modes of MGA as well as by TRIFID and by PC-FRAM. The BEGe detector, because of its good low energy resolution characteristics combined with a medium efficiency at higher energies, was also set to use a gain of 0.075 keV/channel. However, instead of using 4096 channels, we used 16384 channels to allow the same data to be analyzed using MGA in both low and high-energy modes as well as using TRIFID and PC-FRAM.

The shaping time for each of the systems was selected to be representative of a system that would be used in actual inspections, where the samples would potentially have a wide range of activities. For the Inspector-2000 MCA and the DSA-2000 MCA systems, which are both DSP-based systems, we chose a rise time to be 5.6 ms, and the flat top to be 0.8 ms. For the Inspector MCA, which has analog signal processing electronics, we chose a 2 ms shaping time. This setting is generally considered a good compromise between obtaining a good resolution while also providing a good dynamic count rate range. The resolution for the LEGe measurements varied approximately between 530 eV and 550 eV at 122 keV. The resolution for the coaxial measurements varied between 900 eV and 1000 eV at 208 keV. The resolution for the BEGe measurements was approximately 610-660 eV at 122 keV, 710-790 eV at 208 keV.

For the LEGe measurements with the Inspector-2000 MCA, we chose a sample to detector distance between virtually zero and about 20 cm, and typically two layers of a tin filter (a total of 1.6 mm) in front of the detector that kept the dead time at or below 10 percent. In actual practice, the dead time varied between about 0.1% and 13.5%. In addition, samples R and S were measured with 0.8 mm tin filters. To obtain reasonable statistics, we counted the samples until we had approximately 10^6 counts in the spectrum. For most of the samples, it was sufficient to acquire the spectrum for 5 minutes (300 seconds). A few were counted somewhat longer, including some for 400 seconds, 600 seconds or 1000 seconds. Samples P and Q were counted for 40,000 seconds.

For the coaxial detector measurements with the Inspector MCA, we followed similar guidelines. The goal was to keep the dead time reasonable, and to end up with a sufficient amount of counts in the spectrum for decent statistics. Due to the higher efficiency of the coaxial detector compared to the LEGe detector, and the difference in the electronics, the sample to detector distances varied from about 2 cm to as much as 35 cm. We used cadmium as a filter to reduce the count rate. Most measurements were made with 1mm of cadmium, although a few were also made with 0.5 mm of cadmium. To keep the count times somewhat reasonable, we allowed the dead time to be approximately 20%. Most of the samples varied between 15% dead time and 24% dead time. Samples P and Q had dead times in the range of 5% and sample N a dead time of approximately 43%. With these dead times, most samples were counted for 5 minutes (300 seconds), although a few were counted either 400 seconds or 600 seconds. Sample P was counted for 10,000 seconds and sample Q for 40,000 seconds.

For the BEGe measurements with the DSA-2000 MCA, we also tried to keep the dead time reasonable, and the statistics good enough to draw some conclusions from the results. The sample to detector distance varied between 2 cm and 35 cm. Except for samples R and S, all measurements were made with a 1 mm cadmium filter. The count time was again mostly 5 minutes (300 seconds), although a few of the spectra were measured somewhat longer, such as 400 seconds or 600 seconds. Sample P was measured for 10,000 seconds. The dead times varied between about 15% and 25%, except for sample P, where the dead time was about 2.3%.

The raw spectra from the LEGe measurements were used as is for the traditional MGA analysis. The other spectra were digitally manipulated in such a way that we were able to perform the analyses on them. The coaxial detector spectra were manipulated into 4K low energy spectra with a 0.075 keV/channel gain setting and analyzed with MGA in its traditional low energy mode. The same spectra were also manipulated to produce a 4K spectrum with a 0.25 keV/channel setting and analyzed with MGA in the high-energy only mode and with TRIFID. The original 8K spectra were analyzed with FRAM. The BEGe spectra were also digitally manipulated into 0.075 keV/channel 4K spectra and analyzed with the traditional low energy mode of MGA. In addition, the BEGe spectra were manipulated into 4K 0.25 keV/channel spectra, which were analyzed by the MGA high-energy only mode, as well as by FRAM and TRIFID. We used MGA V9.63, FRAM V3.4 and TRIFID V92.4 for our analyses.

All analyses were performed both with the default calculation for the ^{242}Pu weight percentage, and later when the known ^{242}Pu results were made available, with the known ^{242}Pu results as input. All results were decay corrected to January 1, 2000, which was the date of the declared values. These decay corrected results were compared to each other as well as to the declared values that were made available after the first set of results was turned in.

RESULTS AND DISCUSSION

Generally, it seemed that all three isotopics codes produce good results, when they do produce results at all. A few of the samples had odd plutonium compositions that could not be analyzed with the codes without resorting to special manipulations. We purposely did not do so, assuming that an average user of these types of systems would either not be knowledgeable enough to do so, or would not be able to spend the time on such a thing in the field.

Of all the codes, the traditional low energy MGA results seemed to provide the smallest deviation from the known isotopics. An example of the results with such an analysis mode for the LEGe detector spectra and the manipulated BEGe detector spectra for ^{239}Pu are shown in Figure 1. The results are shown for a scenario where the ^{242}Pu was calculated by inference from the other isotopics. The root mean square (RMS) of the percent deviations of the MGA LEGe results from the known ^{239}Pu is about 0.6. Very similar results are obtained with MGA for the BEGe detector in the low energy analysis mode (See Table 1). A comparison between the two types of detectors when both sets of spectra are analyzed with the low energy MGA method is shown in a different manner in Figure 2.

Table 1: A summary of relative percent deviations from known weight percentage values for ^{239}Pu.

Analysis Method (Detector)	Unknown ^{242}Pu		Declared ^{242}Pu	
	RMS Deviation	Chi-Squared	RMS Deviation	Chi-Squared
MGA Low Energy (LEGe)	0.62 %	34	0.43 %	35
MGA Low Energy (BEGe)	0.66 %	27	0.32 %	23
MGA High Energy (Coax)	3.09 %	11	2.90 %	11
MGA High Energy (BEGe)	4.19 %	15	3.81 %	13
FRAM (Coax)	1.32 %	14	1.31 %	21
FRAM (BEGe)	1.81 %	50	1.74 %	62
TRIFID (BEGe)	1.69 %	25		

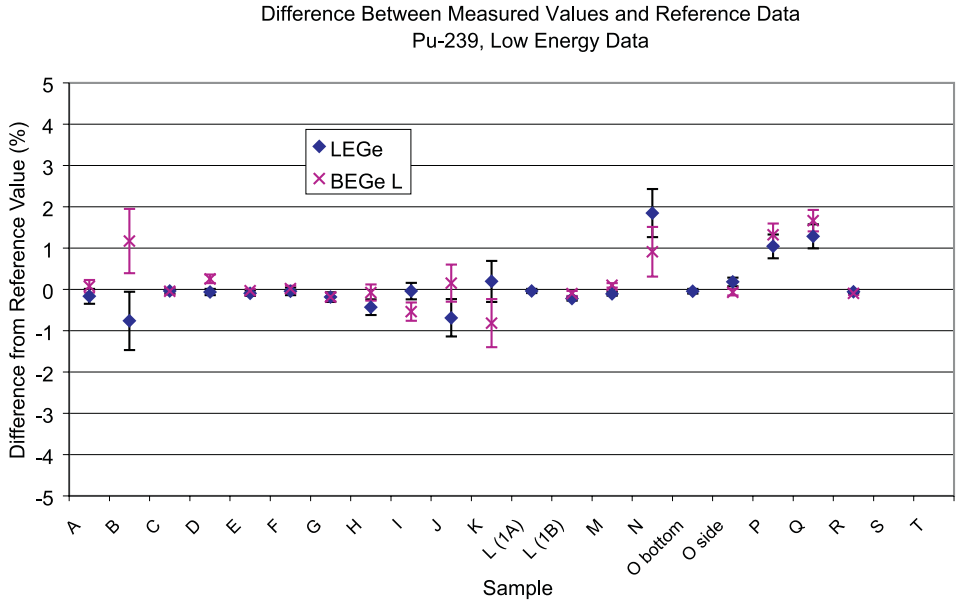

Figure 1
Comparison of measured results with known results with traditional low energy MGA analysis with a LEGe detector and a BEGe detector

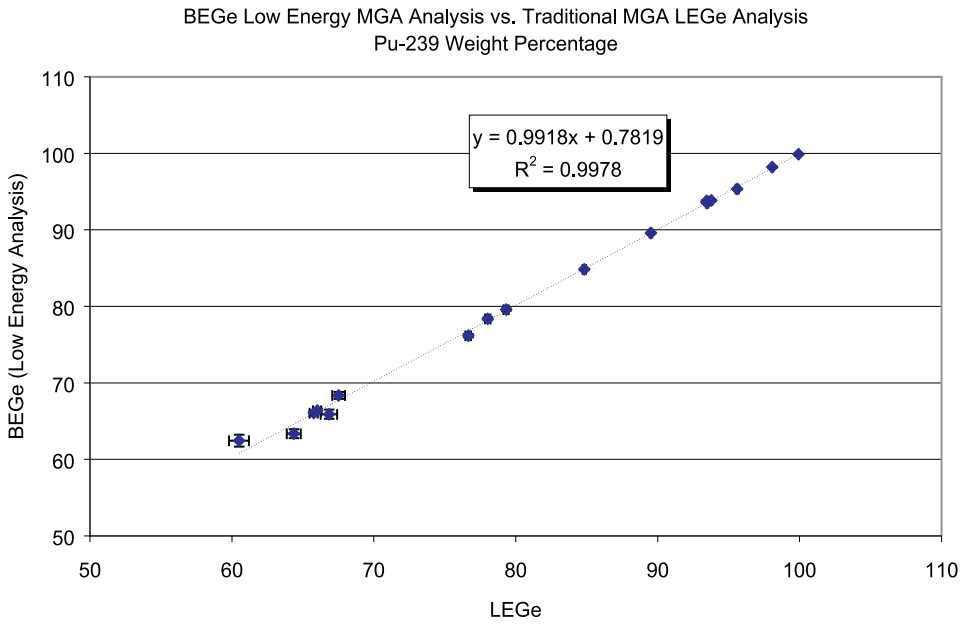

Figure 2
A comparison of MGA low energy results for the LEGe and BEGe detectors

A comparison between the low energy MGA mode and the three methods that use the high-energy mode, i.e. MGA itself in the other mode, FRAM and TRIFID reveals some differences. The RMS of the percent deviations from the known ^{239}Pu value for the BEGe detector and coaxial detector data using the high-energy only mode of MGA are about 4 and 3, respectively. A further investigation reveals that most of the deviation for the high energy BEGe data is caused by a large deviation in the results for sample N. All other results seem to be reasonably close to the correct values. The results from FRAM and TRIFID all show somewhat comparable RMS deviations.

The results for analyses performed with declared ^{242}Pu value show similar behavior. The analysis results using the declared ^{242}Pu value are also shown in Table 1. A comparison of the FRAM results with the BEGe detector against the low energy MGA results with the LEGe detector for the unknown ^{242}Pu case are shown graphically in Figure 3. A comparison of the TRIFID results for the BEGe detector and the low energy MGA results are shown in a similar manner in Figure 4.

Please note that TRIFID is quite capable of analyzing coaxial detector data. We have included only the BEGe detector results in this paper because we were particularly interested in how data from such a detector analyzed. All three methods have been extensively tested with coaxial detectors in the past. The BEGe detector is a new detector type with some unique characteristics. It has potential for plutonium isotopics measurements, if we can show that its performance is acceptable. Evaluating the BEGe detector was one of the specific goals of the Canberra team in this exercise.

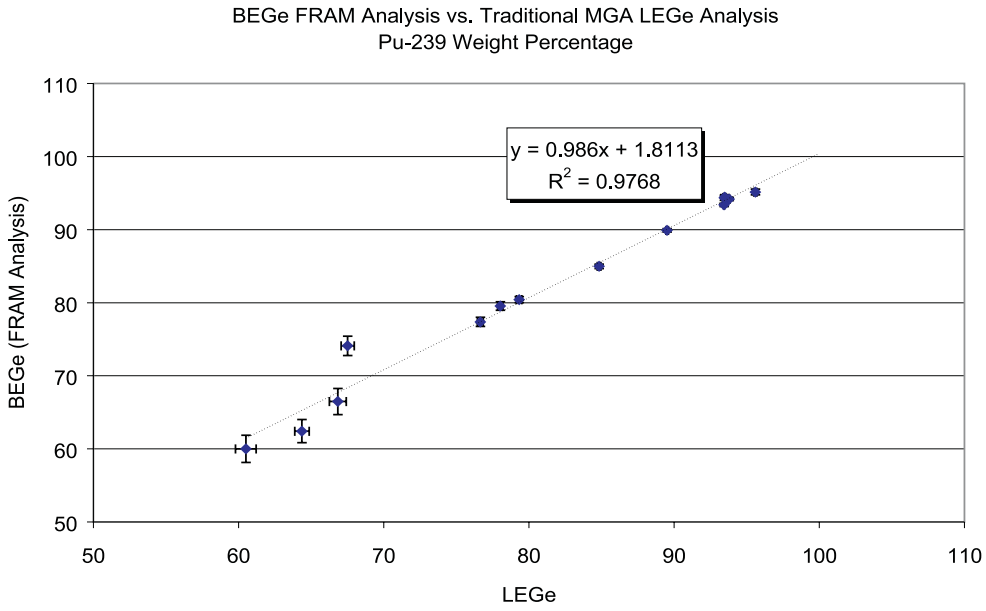

Figure 3
A comparison of FRAM BEGe results and the MGA LEGe results

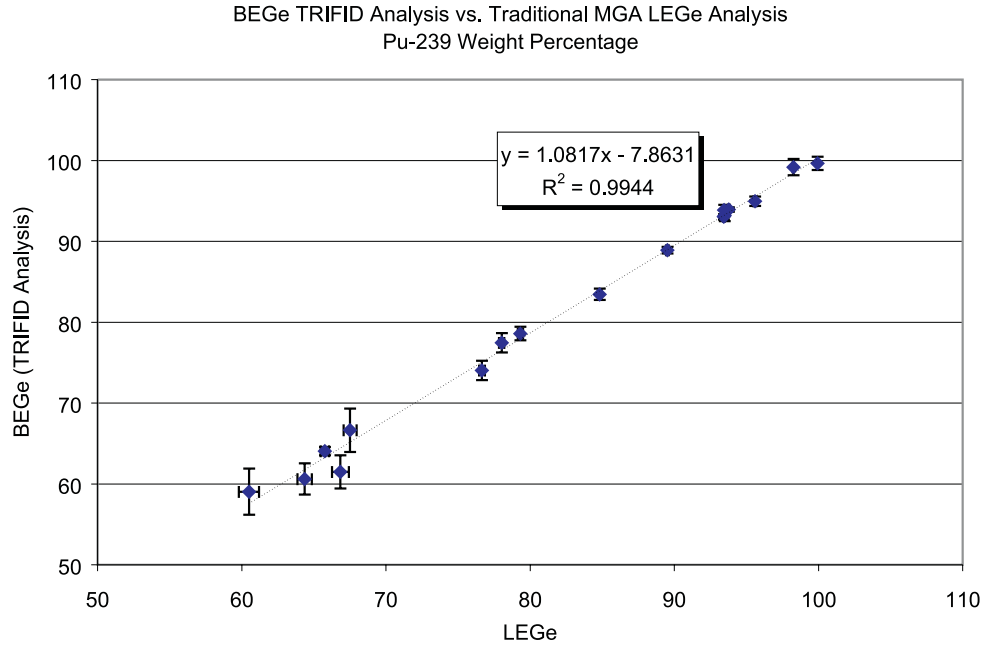

Figure 4
A comparison of TRIFID BEGe results and the MGA LEGe results

The small RMS deviations would seem to imply that the low energy MGA method used with the LEGe detector is the most accurate method to use. However, one must keep in mind that the statistics are not quite the same between each of the analysis methods. Our aim was to keep approximately the same total number of counts in each full energy spectrum – irrespective of the energy region used in the analysis. For the analysis modes that do not use the low energy part of the spectrum, many of the counts in the spectrum do not benefit the analysis since we did not completely block out the low energy part of the spectrum using a filter. The low energy counts dominate the total input count rate from the sample. This results in poorer counting statistics in the high-energy region than would ideally have been used. Usually, the way similar counting statistics are achieved for the higher energy gamma rays is to put in a thin absorber to take out the lower energy gamma-ray contributions (60 and 100 keV region) and then move the detector closer such that the throughput rate for the gamma rays of interest is maximized. We recognize this as a weakness in our test setup. However, it was the fact that we did not block out the low energy region in the spectrum that allowed us to collect spectra that could be analyzed in so many different ways in a reasonable amount of time.

As a result of our choices in choosing the count times for the various spectra, the statistics for the different analysis modes are not equal. We would expect the reported uncertainties of both low energy MGA cases to be the lowest, the reported uncertainties from FRAM and TRIFID somewhat higher, and the reported uncertainties from the high-energy only mode of MGA to be the highest. This is because the "high energy" region used in the analysis in FRAM and TRIFID is not the same as the "high energy" region used by the high energy only mode of MGA. The former start the analysis at 120 keV, while the latter starts at 200 keV. This makes a significant difference in the statistics of the peaks. The results confirm this expected behavior. Both low energy MGA cases have approximately equal statistics and the reported ^{239}Pu weight percentage relative error was below 1 percent for all test cases. The range of the relative errors for the FRAM analyses is about 0.2 – 4.6 and for TRIFID is about 0.3 – 4.8. The MGA high-energy only results show a variation in the reported ^{239}Pu weight percentage relative error from about 1 percent to as high as 27 percent.

To evaluate if the results are reported with appropriate error bars relative to how large the deviations from the correct numbers are, we calculated the chi-squared value for each of the analysis methods. The chi-squared value for the ^{239}Pu analyses is also shown in Table 1. Note that the chi-squared values have been calculated by including only the results for those samples that could be analyzed by all methods. There were several samples that could not be analyzed at all by one method or another. This left us with a total of 14 samples for this test, which gives us 14 degrees of freedom. The acceptable range of chi-squared values at 95% confidence interval for 14 degrees of freedom is approximately between 6 and 26, and at 99% confidence interval about 4 to 31.

We did a similar analysis of the reported deviations and chi-squared values for the other plutonium isotopes. The results for ^{240}Pu are summarized in Table 2, for ^{241}Pu in Table 3, for ^{242}Pu in Table 4, and ^{238}Pu in Table 5.

It can clearly be concluded from the different RMS deviations that are shown in these tables that all three isotopics codes provide results that are very close to the correct results. It can also be concluded that the uncertainties reported by the different codes do not pass the chi-squared test for all isotopes. Many of the analysis/detector combinations actually fare very well, but passing the chi-squared test for everything should not be expected. The uncertainties reported by the codes are derived primarily from the uncertainties in each of the peak areas that are used to determine the isotopic weight fractions. Any systematic uncertainties, e.g. due to the attenuation of gamma-rays in the source/capsule that differs from the assumptions in the physical model, which all of the codes have to rely on.

Table 2: A summary of relative percent deviations from known weight percentage values for ^{240}Pu.

Analysis Method (Detector)	Unknown ^{242}Pu		Declared ^{242}Pu	
	RMS Deviation	Chi-Squared	RMS Deviation	Chi-Squared
MGA Low Energy (LEGe)	0.49 %	39	0.42 %	38
MGA Low Energy (BEGe)	0.30 %	24	0.30 %	24
MGA High Energy (Coax)	3.05 %	10	3.11 %	10
MGA High Energy (BEGe)	4.37 %	14	4.14 %	14
FRAM (Coax)	1.45 %	21	1.40 %	20
FRAM (BEGe)	2.04 %	65	2.00 %	64
TRIFID (BEGe)	1.77 %	16		

Table 3: A summary of relative percent deviations from known weight percentage values for ^{241}Pu.

Analysis Method (Detector)	Unknown ^{242}Pu		Declared ^{242}Pu	
	RMS Deviation	Chi-Squared	RMS Deviation	Chi-Squared
MGA Low Energy (LEGe)	0.047 %	23	0.047 %	28
MGA Low Energy (BEGe)	0.050 %	47	0.047 %	49
MGA High Energy (Coax)	0.185 %	17	0.172 %	17
MGA High Energy (BEGe)	0.299 %	26	0.283 %	26
FRAM (Coax)	0.087 %	87	0.081 %	92
FRAM (BEGe)	0.328 %	437	0.345 %	510
TRIFID (BEGe)	0.089 %	32		

Table 4: A summary of relative percent deviations from known weight percentage values for ^{242}Pu.

Analysis Method (Detector)	RMS Deviation	Chi-Squared
MGA Low Energy (LEGe)	0.70 %	148
MGA Low Energy (BEGe)	0.73 %	164
MGA High Energy (Coax)	0.55 %	62
MGA High Energy (BEGe)	0.57 %	89
FRAM (Coax)	0.95 %	29
FRAM (BEGe)	0.88 %	27
TRIFID (BEGe)	0.55 %	59

Table 5: A summary of relative percent deviations from known weight percentage values for ^{238}Pu.

Analysis Method (Detector)	Unknown ^{242}Pu		Declared ^{242}Pu	
	RMS Deviation	Chi-Squared	RMS Deviation	Chi-Squared
MGA Low Energy (LEGe)	0.013 %	36	0.013 %	40
MGA Low Energy (BEGe)	0.018 %	50	0.017 %	55
MGA High Energy (Coax)	0.135 %	52	0.130 %	52
MGA High Energy (BEGe)	0.272 %	90	0.120 %	90
FRAM (Coax)	0.018 %	8	0.009 %	8
FRAM (BEGe)	0.087 %	85	0.092 %	109
TRIFID (BEGe)	0.028 %	34		

Likewise, the ^{242}Pu is determined using an empirical formula, and if the model produces a bias in ^{242}Pu, it has an effect on all the other isotopics as well, because the weight percentages have to add up to 100. Since the magnitude of these types of systematic uncertainties is sample dependent, the chi-squared value can easily become elevated. Remarkably, the chi-squared values are quite good for many of the analysis/detector combinations. But, there seems to be at least one or two isotopes for which each code produces a high chi-squared value. For example, MGA and TRIFID show elevated chi-squared values for ^{242}Pu. FRAM shows a high chi-squared value for ^{241}Pu.

It should be pointed out that of the three programs, FRAM needs the most operator intervention to produce its results. It seems to require the energy calibration to be very correct before producing an analysis result. The other two codes are more forgiving in this regard. It also does not automatically detect the presence of uranium in the MOX samples, unlike the other codes. And, FRAM requires the operator to manually intervene to decay correct the results to the declaration date.

CONCLUSION

The Pu-2000 inter-comparison exercise organized by the ESARDA NDA Working Group, and Dr. Michael Bickel in particular, has permitted us to measure a set of plutonium samples to test several different hardware configurations. The data we collected has permitted us to analyze the spectra using three commonly used plutonium isotopics codes. There were some samples, for which not all programs and setups produced an answer. We are still evaluating the reasons for such failures, but we suspect that it was primarily due to insufficient statistics. We have calculated the root mean differences from the known values for all the results that each of the analysis methods and detector configurations produced. In general, all three codes seem to provide results that are very close to the correct values, although there clearly are some small differences between the different methods.

The BEGe detector is clearly a viable candidate to make these kinds of measurements, not only in the high-energy mode, but also for the MGA low energy analyses.

Since all the codes seem to produce reasonably good results, if someone obtains poor results, it may have more to do with the operator's familiarity of the specific code and personal preferences. This is, in fact, the premise why Canberra has deemed it prudent to obtain licenses for all three codes.

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