Achievements on Nondestructive Assay Isotopic Enrichment Measurement Systems under the USDOE/NNSA-ABACC Safeguards Cooperation Agreement

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ABSTRACT

The United States Department of Energy/National Nuclear Security Administration (DOE/NNSA) and the Brazilian-Argentine Agency for Accounting and Control of Nuclear Materials (ABACC) agreed to cooperate on the testing and evaluation of special nuclear material isotopic measurement software. The project is significant because it provides a regional forum for technical experts to evaluate and standardize isotopic enrichment measurement software used to verify operator declarations for enriched uranium. The project was initiated by establishing an Isotopics Measurements Working Group (IMWG). The goal of the Group is to test existing software packages and to provide guidance for standardizing and upgrading the software to broaden the applicability, improve the data quality, and ensure that software quality assurance objectives are being met. Starting in 2006 the IMWG members have collected over 7500 uranium spectra using defined hardware systems and experimental environment. Collected data have been analyzed with software packages that use enrichment meter, peak fitting and peak ratio methods for uranium enrichment analysis. This paper describes the results obtained by the IMWG and provids details on the methods and procedures used for testing and evaluating the performance of the various software packages. Recommendations for standardization and improvements of analysis methods and algorithms are provided.

INTRODUCTION

In 1994, DOE/NNSA and ABACC signed a technical cooperation agreement on research, development, testing, and evaluation of technology, equipment, and procedures in order to improve international nuclear materials safeguards technologies in South America. As a part of this agreement, under Action Sheet 14 (AS14), which was signed on April 18, 2005, ABACC and DOE agreed to cooperate on the testing and evaluation of uranium isotopic measurement software. Currently, ABACC and DOE are using various versions of several software packages for uranium isotopic abundances determination. Inconsistencies between the versions of the software have resulted in data discrepancies that often have to be manually corrected. The goal of AS14 is to evaluate the variety of isotopic software analysis packages that are currently used by DOE and ABACC to verify operator declarations for enriched uranium and provide guidance for standardizing and upgrading the software to broaden the applicability, improve the data quality, ensure that software quality assurance procedures are being met, and control the final product's distribution.

To accomplish the objectives of AS14, ABACC and DOE established the Isotopics Measurements Working Group (IMWG). The scope of the IMWG is identification, testing, and evaluation of available gamma spectroscopy analysis codes used to determine the isotopic abundances of uranium samples. The tests and evaluations will identify the strengths and weaknesses of available codes and will result in a set of recommendations for improving and standardizing software used to evaluate uranium spectra. The results of the study will provide guidance for development of a standard uranium spectral analysis software package that better meets the needs of the end user and is consistent with regional and international quality assurance requirements for the safeguards community.

The IMWG devised effective test plans that provided uniformity in the evaluation processes for each software package. Specific hardware systems, software packages, source material standards, methods, and procedures for evaluation of analysis codes were identified at each participating laboratory. The tests goal was to evaluate agreed-upon attributes for the various types of software that are being used to determine isotopic abundances for uranium. Tests and evaluations were conducted for two analysis techniques: 1) uranium enrichment and 2) uranium isotopics. Software packages that use the enrichment meter, peak fitting, and peak ratio methods were evaluated for the uranium enrichment technique. The IMWG uses peak ratio method software for evaluation of uranium isotopics.

According to the test plans, each laboratory was primarily responsible for collecting using specific hardware configurations and for testing the software packages listed in Table 1. Individual laboratories were also asked to analyze spectra collected at another laboratory for codes evaluation results verification.

Details on the AS14 objectives, scope, structure and organization of the IMWG, and initial test plan were presented at the 47th INMM Annual Meeting in 2006[1].

Table 1. Software to be tested by each laboratory for the first test plan

	ORNL	**LLNL**	**LANL**	**ABACC**
Enrichment meter method	WinU235			Win U235 WinUF6 MGAU
Peak fitting method	NaIGEM	--	--	NaIGEM
Peak ratio method	U/Pu, MGAU	MGA++,U235HI	PC-FRAM	--

TEST PLAN

The work at the laboratories supporting the IMWG have initiated implementation of their test plans. The status for the first round of testing for each laboratory is provided below.

The test plan for LLNL utilized three HPGe systems, two different ORTEC MCAs, three different sets of standards, and various thicknesses of aluminum and stainless steel absorbers. LLNL has collected and analyzed more than 1000 spectra using 0.075keV/channel gain at optimal geometry. The spectra were saved in both .spc and .spe formats.

The primary purpose for the LANL measurements was to evaluate the performance of the software codes as a function of peak resolution and peak shape. LANL set up the data acquisition systems to obtain spectra that vary in peak shape characteristics (e.g., variations in the FWHM or resolution and variations in the low tails of the peak). LANL used two HPGe detector systems (one planar and one coaxial), the ORTEC DSPEC Plus MCAs, and eight sets of standards. Variations in the rise time of the DSPEC Plus from 0.2 up to 8.0 µs were made to obtain spectra with various peak shape characteristics. In order to obtain spectra with various shapes, they used a rise time of 4.0 µs and manually adjusted the PZ to produce peaks with low-energy tails of various sizes.

ORNL conducted measurements with two sets of standards (low enrichment: 0.31 wt.% to 4.46 wt.% ^{235}U and high enrichment: 20.11 wt% to 93.17 wt.% ^{235}U). The instrumentation configuration included four NaI, one LaBr3 and two HPGe (one planar and one coaxial) detectors systems in various combinations with the following MCAs; MCA-166, ORTEC® DART, ORTEC® digiDART, Canberra InSpector 2000, and Canberra InSpector 1000. A set of aluminum, iron and monel absorbers were used to simulate various container materials. More than 4300 spectra were collected.

ABACC conducted measurements with three NaI detectors system, three HPGe (planar) detectors systems and one CZT system (1500 mm^3 detector), the MCA-166 – GBS Elektronik MCA, one set of SRM-969 standards, and a set of aluminum, iron and monel absorber materials. Spectra acquisition time was set to 1000 seconds and 10000 seconds for calibration purpose. More than 1000 spectra have been collected and analyzed.

TEST PLAN RESULTS AND DISCUSSION

Once individual laboratory tests were complete, the IMWG members met at the ABACC Headquarters in Rio de Janeiro, Brazil on February 28th 2007. The purpose of the meeting was to compile all results from the tests and coordinate analysis and evaluation of the test results. The results of the application of listed above software packages for analyses of the spectra obtained in the individual laboratories are shown bellow.

ABACC compared the results of WinU235 and NaIGEM software for the spectra obtained with NaI detectors. NaIGEM showed better results than WinU235, but for both programs the input value of density or attenuation coefficient for the absorbers had to be modified in order to achieve better calculated enrichment values. This is an important point to take into account when measuring the enrichment of an item with high attenuation. The effect of the attenuation is more sensible to the WinU235 that uses the enrichment meter method compared to the NaIGEM that uses the peak fitting method. ABACC utilized the program ATEN (developed by Cristallini;O.- ABACC) to calculate the "apparent" density or attenuation coefficient. The comparison of the WinUF6 (v.1.00.0014) and MGAU (3.2; 3.21; and 4.0) software for spectra obtained with HPGe detectors showed minor differences between the programs results for the enriched meter method.

ABACC identified several issues regarding use of the enrichment meter method codes. These issues include development of best practice procedures to minimize errors in the field, propagation of total uncertainty in the codes and incorporation of correction factors in the bias observed when using the WinU235 code for thicker absorber material. These issues need to be resolved because field measurements typically rely upon a single, short count for facility declaration verification (e.g., field measurement conditions are much different from those observed in the laboratory). In addition, it is typically not possible to meet advertised international target values (ITVs) for portable measurement systems. One conclusion is that calibrating the system with absorbers (closer to the real item to be measured) improves the results when measurements are performed in the field. It is recommendable to develop a set of primary traceable absorbers that are an exact match in composition and thickness for cylinders that are certified to store UF_6 (e.g., 30B, 48G, etc.).

In addition, ABACC discussed the need for validating the errors reported in the codes because it is not clear what they include and whether all of the contribution from the uncertainty budget is accounted for properly. ABACC support laboratories performed an uncertainty study using NaI detectors with WinU235 and NaIGEM. The purpose of the study was to determine if the uncertainty reported by the software codes is representative of what is expected from multiple measurements of the same standards. Calibrations were performed using U standards and attenuator materials as well. A comprehensive statistical uncertainty evaluation that included contributions from counting statistics (random), uncertainty in the calibration coefficients (systematic), absorbers (systematic) and wall thickness (systematic) was performed. The study concluded that the total measurement uncertainty is dominated by counting statistics (random) if the time of counting and the overall efficiency of the system is low (typical of field safeguard application). For longer counting time (as in the calibration procedure) systematic errors are predominant. The results indicate that the uncertainty reported by the codes does estimate the uncertainty for counting statistics and calibration coefficients but does not incorporate uncertainties for variations in absorber density or thickness or have inadequate method of analysis. Both codes have to improve their uncertainty calculation.

ORNL analyzed the spectra obtained according to its test plan but did also analyze the ABACC spectra to confirm the issues observed by ABACC. The ORNL analysis confirmed the ABACC results and issues related to the precision of the analysis when thicker absorbers are used, and the poor results presented by WinU235 compared to NaIGEM. ORNL results showed minimal differences for individual data points compared to ABACC results. ORNL provided ABACC with the spreadsheet so that this issue can be further investigated by both sides. There were also minor differences between MGAU 3.21 and MGAU4.0 using the peak ratio analysis method with ORNL data. ORNL will conduct an additional evaluation to determine if revisions to the MGAU 3.21 code to correct for issues associated with reporting the ^{234}U concentration resulted in the reported differences between these two versions of MGAU. In addition, the ORNL studies concluded that the enrichment meter method provides better precision for count times that are consistent with field measurements conducted by ABACC. ORNL data demonstrated, as expected, that the measurement uncertainty increases as the thickness and/or density of absorbers increases. Recommendations from

ORNL evaluations include incorporation of a drop down menu for UF_6 cylinder types in addition to a drop down menu for material types. In addition, it was recommended that the working group devise a plan to utilize modeling (i.e. MCNP) to test and incorporate into or compare with the code.

LLNL analyzed the ABACC spectra data using the peak ratio method that is currently provided in the commercially available MGA++ version 1.06 code from ORTEC. It should be noted that the data collected by ABACC was intended for enrichment meter principle evaluation, and may not be optimal for peak ratio method evaluation due to poor counting statistics in the low energy region utilized by MGA++ for estimating the isotopic concentration for uranium. The analysis conducted by LLNL determined that the U235 subprogram performs inadequately when the statistics are poor. The results generated by the U235 subprogram had also shown a bias when compared to the declared values for the standards. Technical experts at LLNL will investigate this issue. However, improvements were noted in the results when the same spectra were analyzed using an in-house code (MGA4U) that is not currently commercially available. Additional analysis will be conducted on the ABACC spectra using the commercially available MGA++ and MGA4U codes to complete all aspects of the LLNL test plan. It was proposed that MGA4U be the first code to run through an agreed upon standardized test and evaluation process prior to commercial release. In addition, it was concluded that the instrumentation utilized for the peak ratio method should be optimized (i.e., uncollimated detector, longer count times, maximized count rates, etc.). For example, it was determined that the Canberra U/Pu low energy germanium (LEGe) detector is not optimally designed for use with the peak ratio method because of the presence of the internal tungsten collimator that cannot be removed.

LANL presented results from the analysis of four sets of data. The data sets were generated by the LANL, the LLNL, the ORNL and the ABACC. The analyses were conducted using the peak ratio method incorporated into FRAM 4 that is commercially available from Canberra and ORTEC. The analyses concluded that the U120_1001COAX parameter set produces small biases in the results across all enrichments for high statistics data. In addition it was noted that the current commercial version of FRAM permits users to analyze data with different parameter sets. The developer indicated that the next version of FRAM will have strict administrative controls incorporated into the code to prevent the inspector to change parameter sets in the field. It was also determined that the errors reported by the code are constrained to random errors from counting statistics. This represents the precision of the measurement well. However, the user is responsible for evaluating contributions from systematic errors to estimate the total measurement uncertainty. It was determined that FRAM shows inadequate results due to poor counting statistics from depleted and natural uranium data sets collected by ORNL and ABACC that were intended to be analyzed using enrichment meter methods. However, it was determined that the results could be improved if the full-width-at-half-maximum (FWHM) is fixed using the spectrum from the 4.46 wt. % ^{235}U standard. [Note: The FWHM is fixed by checking this option in the analysis parameters for the code.] The FRAM code successfully analyzed all the depleted uranium (0.31 wt. % ^{235}U) and natural uranium (0.71 wt. % ^{235}U) data with the FWHM fixed. Recommendations include implementing administrative privilege so that users cannot arbitrarily alter parameter sets during the analysis. Another recommendation is to include the systematic uncertainty in the next version of the code.

CONCLUSIONS AND RECOMENDATIONS

The IMWG members agreed with the following conclusion statements and recommendations at the meeting held in ABACC headquarters for the first test plan:

- **Conclusion**: The quality of a single, short count time measurement is dependent upon all elements of the measurement process (i.e., software, hardware, personnel qualifications, environment, etc.).The isotopic analysis codes are only one element of many others that need to be controlled in order to achieve a high quality measurement under the constraints that ABACC inspectors often see in the field.
- **Recommendation:** Address comprehensive measures necessary to achieve quality data using portable measurement systems in a field environment.
- **Conclusion:** The uncertainties reported in the codes, in particular WinU235, appear to be inaccurately propagated.
- **Recommendation:** ABACC and ORNL shall conduct a simple study to determine if errors associated with counting statistics are accurately calculated within the enrichment meter method and peak fitting method codes. Results of the study will be provided to the code developers.
- **Conclusion:** The WinU235 code generates biased results when compared to declared values across the range of 0.3 wt. % to 4.46 wt. % ^{235}U.
- **Recommendation:** The system should be calibrated using attenuators to minimize the bias. The fabrication of a primary traceable set of attenuators that matches the thickness and material of construction for cylinders that will be measured in the field would also improve the results of measurements.
- **Recommendation:** The enrichment meter codes should have the capability to select UF_6 cylinder types from a drop down menu within the code that is in addition to selecting default material types.
- **Conclusion:** The ORNL analysis of the ABACC data confirmed the ABACC results in all cases and minor variations in individual data points.
- **Recommendation:** ORNL and ABACC will further evaluate differences in the individual data points to determine the cause for the minor differences.
- **Conclusion:** MGAU version 3.21 produces different results than version 4.0 when the data is analyzed using the peak ratio method.
- **Recommendation:** Follow up with code developer to determine if changes made to the code to correct for errors in the ^{234}U correction impacted results for the low energy region used to analyze for isotopic abundance of ^{235}U and ^{238}U. Additional studies may be necessary to quantify the impact of the revisions to the code results.
- **Recommendation:** The use of modeling codes, such as MCNP, should be evaluated to determine optimal calibration and measurement conditions and to validate changes to the codes. This would limit the number of empirical studies typically needed to test each code.
- **Recommendation:** Utilize the international IMWG to control version distribution of these codes.
- **Conclusion:** The enrichment meter method provides better measurement precision compared to the peak ratio method for measurement conditions commonly seen in the field by ABACC inspectors.
- **Recommendation:** This result is not unexpected because the enrichment meter method is dependent upon the single, highest intensity, higher energy peak for ^{235}U. The final report

for this study should include limitations on the application of each method.

- **Conclusion:** The U235 subprogram for the commercially available version of MGA++ failed to adequately analyze many of the ABACC and ORNL spectra due to poor counting statistics in the low energy region.
- **Recommendation:** The results were improved when the ABACC spectra were analyzed using MGA4U. The MGA4U code should be the first code to run through an internationally approved acceptance standard and certified for commercial release. Revisions to the code should be controlled. In addition, the code developer should be provided with the minimum amount of funding necessary to sustain, maintain and improve the code as new challenges for spectral analysis of fuel mixtures become necessary.
- **Conclusion:** When the U120_1001COAX parameter set is selected the commercially available version of FRAM 4 does not adequately analyze the ABACC and ORNL spectra of depleted and natural uranium with thick absorbers that are commonly generated by portable spectroscopy systems. The code failed to confirm the declared enrichment for many depleted and natural uranium spectra using this parameter set due to poor counting statistics in the analysis region.
- **Recommendation:** Fixing the FWHM parameter improves the capability of the codes to perform under these measurement conditions.
- **Conclusion:** The FRAM 4 code generates a small negative bias in the results across all range of enrichments.
- **Recommendation:** The code developer will investigate the reason for the small bias reported by the code and provide feedback on this issue to the IMWG.
- **Conclusion:** The current version of the FRAM code permits the user to select variable parameter sets. This could lead to errors in the analysis of the spectral data.
- **Recommendation:** The developer will incorporate administrative restrictions within the software that prevent the user from changing parameter sets in the field. This new version will also be tested and evaluated using an internationally acceptable set of standards prior to commercial release. Revisions to the code should be controlled. In addition, the code developer should be provided with the minimum amount of funding necessary to sustain, maintain and improve the code as new challenges for spectral analysis of fuel mixtures become necessary.
- **Conclusion:** The current version of the FRAM code presents only random errors associated with counting statistics.
- **Recommendation:** Revisions to the code should include estimates for systematic uncertainties. The new version of the code should be tested in accordance with the recommendation mentioned above.
- **Conclusion:** The current enrichment determination codes lack sufficient diagnostic capabilities to address limitations for the codes.
- **Recommendation:** The IMWG should define a set of minimum diagnostic requirements for the various versions of the codes and work with code developers to incorporate these diagnostic capabilities into the codes.
- **Conclusion:** Many of the issues related to sustainability, version control and standardization for AS 14 were also identified as issues within the ESARDA NDA working group.
- **Recommendation:** Utilize the experience of the ESARDA NDA working group to supplement AS 14 activities and expand the IMWG to an international forum.

- **Conclusion:** The international safeguards community needs to develop internationally accepted standards or best practice guides to standardize methods for testing these codes and providing information on the use and limitations of these codes.
- **Recommendation:** Form an international IMWG to improve communication between technical experts on issues related to these codes. The international IMWG should maintain a website and produce a newsletter to keep the international users community informed.

REFERENCES

1. Perrotta,J.A; Frazar,S; McGinnis,Brent "Cooperation on nondestructive assay isotopic enrichment measurement systems under the DOE/NNSA-ABACC safeguards cooperation agreement" ; INMM 47th Annual Meeting, Nashville, July 16-20,2006.