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Sustainability of Gamma-ray Isotopics Evaluation Codes

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Abstract

In November 2005, the international workshop "Gamma Evaluation Codes for Plutonium and Uranium Isotope Abundance Measurements by High-Resolution Gamma Spectrometry: Current Status and Future Challenges" was held in Karlsruhe, Germany. Some of the main issues discussed during the November 2005 meeting were related to concerns voiced by international inspectorate authorities such as the International Atomic Energy Agency (IAEA), the European Atomic Energy Community (EURATOM), and the Brazilian-Argentine Agency for Accounting and Control of Nuclear Materials (ABACC) about the standardization and sustainability of gamma-ray isotopic analysis codes that are commonly used during safeguards inspections. A follow-up international workshop was conducted in Oak Ridge, TN in 2008. This workshop was in response to needs expressed by the international safeguards community during the Karlsruhe meeting and recommendations made under Action Sheet 14; a cooperative effort between the U. S. Department of Energy and ABACC. The purpose of the Oak Ridge workshop was to bring code developers and end users together to better understand the capabilities and limitations of the codes; to discuss mechanisms to ensure these codes are sustained and quality tested; and to ensure updates or revisions are performed in a controlled manner. During an Action Sheet 14 meeting held in Rio de Janeiro, Brazil in which the IAEA and EURATOM participated as observers, and in subsequent meetings of the European Safeguards Research and Development Association (ESARDA), all parties agreed that the regional working group initially established under the DOE/ABACC cooperation should be expanded to an international working group. The purpose of the international working group is to provide a forum to exchange information, discuss technical developments, and validate and test the various codes. However, progress to formally establish the working has been slowed by a lack of dedicated funding and competing priorities within the various international organizations. Recently, the ESARDA Nondestructive Assay Working Group established a dedicated website for the International Working Group on Gamma Spectrometry Techniques (IWG-GST). In this paper, we will explore specific steps that should be taken to strengthen this working group's ability to affect development of a common testing platform and address concerns regarding applicability, sustainability and version control for these important codes.

1. Introduction

In November 2005, the international workshop "Gamma Evaluation Codes for Plutonium and Uranium Isotope Abundance Measurements by High-Resolution Gamma Spectrometry: Current Status and Future Challenges" was held in Karlsruhe, Germany[1]. Some of the main issues discussed during the meeting were related to concerns voiced by international inspectorate authorities such as the International Atomic Energy Agency (IAEA)[2,3], the European Atomic Energy Community (EURATOM), and the Brazilian-Argentine Agency for Accounting and Control of Nuclear Materials (ABACC) about the standardization and sustainability of gamma-ray isotopic analysis codes that are commonly used during safeguards inspections. During an Action Sheet 14 meeting held in Rio de Janeiro, Brazil in which the IAEA and EURATOM participated as observers, and in subsequent meetings of the European Safeguards Research and Development Association (ESARDA), all parties agreed that the regional working group initially established under the DOE/ABACC cooperation should be expanded to an international working group. A proposal to launch The International Working Group on Gamma Spectrometry Techniques (IWG-GST) under the auspices of the ESARDA NDA Working Group was accepted in a meeting organized in Aix-en-Provence, France, in May, 2007. A follow-up international workshop was conducted in Oak Ridge, TN in 2008[4] in response to the Karlsruhe meeting and recommendations made under Action Sheet 14[5]. The purpose of the Oak Ridge workshop was to bring code developers and end users together to better understand the capabilities and limitations of the codes; to further discuss mechanisms to ensure these codes are sustained and quality tested; and to ensure updates or revisions are performed in a controlled manner. In the following, we will first describe the background and recent development of the main enrichment measurement and plutonium isotopics methods and then explore specific steps that are being taken and should be taken to strengthen this working group's ability to affect development of a common testing platform and address concerns regarding applicability, sustainability and version control for these important codes.

2. Current Codes

The current gamma-ray isotopics evaluation codes can be broadly divided into two categories; measurements with medium resolution detectors, such as NaI or CdTe detectors that operate in ambient temperature; and measurements with high resolution detectors, such as HPGe detectors. Medium resolution detector systems based on NaI detectors are used extensively by international safeguards inspectorates from ABACC, EURATOM and the IAEA to verify uranium enrichment [6,7]. This routine use is generally built around the enrichment meter method[8], which uses the 185 keV peak of U-235 as the basis of the analysis. Example portable instruments that use the enrichment meter method include the PMCA[9] and the IMCA[10,11]. High resolution detector systems can also be used for uranium enrichment measurements with the enrichment method[12] and are required for plutonium analysis. Several software packages have been developed for this purpose over the years. The most well known plutonium isotopics codes are MGA[13,14] and FRAM[15]. Codes that are less well known and not as widely used by the international community include TRIFID[16] and IGA[17].

Unfortunately, there are two somewhat divergent codes that both use the MGA name. The original Ray Gunnink MGA code has been commercialized through Canberra, and enhanced for example to deal with poor statistics and problematic peak shapes, to provide high energy only analysis, and to include uranium analysis under the name MGAU [18,19,20,21,22,23]. At the same time, Lawrence Livermore National Laboratory retained a copy of the MGA for plutonium analysis at the time of Dr. Gunnink's retirement and has commercialized it and other related routines described below through Ortec under the name MGA++ [24,25,26]. The distinction between MGA and MGA++ is subtle. The MGA++ is a suite of codes that include an improved MGA for plutonium analysis; a slightly different U235 code than MGAU for uranium analysis; MGAHI, that uses only >200 keV plutonium data for plutonium isotopic analysis; and CZTU - a uranium isotopic analysis via a CZT detector[27]. However, the underlying unfolding engines are mostly using routines originally created by Dr. Gunnink. [28,29].

The FRAM code has also been developed further[30,31,32] at Los Alamos National Laboratory. The latest development added many new features in response to new measurement requirements and to meet user needs [33,34]. Some of the improvements worth mentioning here are:

- Physical model efficiency curve fit in addition to the existing empirical model curve;
- Capability to fit the X-ray peaks;
- Uranium analysis enhancements, which include the corrections for the loss of peak areas due to summing, the ^{236}U correlation prediction, and the decay correction for non-equilibrium $^{238}U/^{234}Th$;
- Capability of isotopic analysis data taken with the Peltier-cooled CdTe detectors [35,36]; and
- Automated parameter file selection to speed the measurements process for robotic applications or applications for samples with largely unknown contents.

Since the isotopics codes were originally conceived, there has been a significant amount of development in detector technology, signal processing electronics and computer technology. The electronics have become digital, and more portable. In most cases, a digital implementation provides superior throughput, but requires a different setup and optimization to make the electronics match the detector and for the isotopics codes to work properly. Advancements in computer processors and changes in operating systems have resulted in updates to various codes.

For medium resolution detectors, LaBr crystals have begun to replace the NaI detectors in many applications due to improvements in resolution. Additional scintillation crystals with comparable or better characteristics are being investigated. This has prompted a renewed interest in using more sophisticated analysis algorithms for these medium resolution detectors[37,38]. The more sophisticated analysis algorithms generally use a peak fitting technique which is an advanced version of the enrichment meter technique that utilizes computed responses that are fit by least squares to the spectral data. Since the technique uses computed spectrum responses to analyze the data, the results are

generally more accurate for the same measurement system and conditions. In addition, this technique can account for interferences from other isotopes such as thorium. The NaIGEM [39] analysis software, also developed by Dr. Gunnink, is one such example. The NaIGEM software is commercialized through ICx Technologies and GBS Elektronik has been approved both by ABACC and the IAEA for safeguards inspections.

Cadmium Zinc Telluride (CZT) is beginning to replace more conventional CdTe detectors. Conventional CdTe detectors are available in larger volumes than CZT but have a lower resistivity and band gap (1.6 eV for CZT compared to 1.47 eV for CdTe)[40]. The higher band gap translates into a lower leakage current and a potentially better resolution. The relative efficiency of the CZT material is also approximately 3 times higher than the efficiency of a CdTe detector at 81 keV and as much as 20 times higher at 356 keV. CdTe detector performance can be improved with the use of pulse risetime discrimination and/or Peltier cooling. Relative to the other detector types, the CZT detectors are still quite small. Many implementations exist where multiple CZT crystals are combined with sophisticated electronics to provide a single composite system with better detection sensitivity than using a single crystal. The miniaturized digital electronics implementations make this more practical and affordable than ever. This has prompted analysis algorithm development with more sophisticated approaches.

HPGe detector manufacturers have developed extended energy range detectors that provide resolution that is similar to the traditional smaller detectors, while providing an acceptable resolution to use the isotopics codes with the added benefit of an increased detection sensitivity relative to the small low energy only detectors[41,42,43,44,45]. However, the setup parameters of the isotopics codes need to be adjusted for optimum performance. In addition, the electrical cooling technology has become good enough to produce an electrically cooled, portable HPGe detector system that can be used for isotopics measurements in the field[46,47] without the use of liquid nitrogen.

Additional challenges are posed to the isotopics codes by their extensive use for new materials and material combinations that were not in use during the original design phase of the codes. This includes measurement of low-activity, heterogeneous waste commonly encountered during cleanup of old nuclear sites. Some of these scenarios are being at least partially resolved with the combination of modeling software and the isotopics codes[48, 49].

Another critical point is related to quality assurance requirements established by national and international regulatory bodies. Users, code developers and vendors have to follow several steps if they want to have their quality system recognized by external organizations. In this regard, elaboration of codes and reporting of the results should be appropriately documented and validated, preferably using an internationally agreed upon methodology. This usually requires a level of transparency that commercial code developers may not want to provide due to the proprietary nature of the code. However, this level of transparency is needed to resolve questions or discrepancies that may arise from results generated by the codes that do not agree with process knowledge or other

analysis techniques. It is important that each type of code provide similar results for similar spectra.

3. Further Development and Sustainability

It is quite clear from the review above that activity to maintain these important codes continues by many parties. At the same time, these efforts are driven by sometimes rather exotic situations only. There is no generally accepted method to disseminate information regarding updates, nor is there a generally accepted envelope of scenarios that the codes are designed to address. The goal of the International Working Group on Gamma Spectrometry Techniques (IWG-GST) is to provide a forum for exchange of this kind of information, technical developments, and validation and testing of gamma-spectroscopy techniques used to determine the isotopic composition of uranium and plutonium samples. The IWG-GST will also address issues related to gamma evaluation codes, such as their applicability, capability and limitations, standardization, sustainability, and version control. Under the working group, technical personnel from the IAEA, national and international research institutions, the detector and instrument manufacturers, and the nuclear fuel cycle industry can provide insight to issues commonly encountered when the codes are used in a laboratory or field measurement environment. The code developers get an opportunity to understand the end user issues to provide information and suggestions to the end users related to the capabilities and limitations of the individual codes.

To facilitate this exchange of information, the IWG-GST has started the development of a testing platform for gamma evaluation codes. This platform, which will be hosted in the webpage of the ESARDA NDA working group[1], will collect a set of gamma spectra useful to test the performance of isotopics evaluation codes. The target users of the platform will be:

- code developers that can use the platform to assess the capability of their newly developed codes or new versions of existing codes; and
- analysts that can use the platform to compare different codes in order to select the one that best fits their needs.

The tentative architecture of the platform has been discussed in several meetings and in principle agreed within the IWG-GST. The test platform will contain different sections with different purposes:

- set of spectra acquired in ideal measurement conditions mostly devoted to performance assessment of the accuracy of codes in a "typical" range of application;
- spectra acquired in non-ideal conditions to test the robustness of the analysis to harsh or bad conditions; and
- spectra acquired in unusual situations in order to test the extension of the application of codes outside the present "normal" operational conditions.

[1] The library of spectra and testing platform can be found at http://esarda2.jrc.it/internal_activities/WG-NDA/index.html. Access to the site is restricted but a password will be provided upon request.

The most relevant part of the platform will be the first section, because this is where the user will be able to derive useful information about the capability, accuracy and limitations of each code. This section will also provide a controlled mechanism for mutual inter-comparison benchmarking of the codes and in the future could be the seed of a possible procedure for code certification. The second and third section can be used to broaden the knowledge about the behavior of codes outside and beyond the normal application conditions and can provide a useful support to the users who need to tackle special applications. The codes are not required to "pass" the tests with spectra of the second and third section, but it will be useful to know the limitations of the code relative to the end users unique measurement application. This will provide the international community with a mechanism for exchanging results for measurements on samples that are not typically encountered in today's nuclear fuel cycle.

Table 1 shows the preliminary list of spectra that will be included in the testing platform. Each spectrum will be documented with an accompanying description file. The spectra will be provided in their native format (.cnf, .chn, .spc, .spe, ...) as well as in an ASCII format in order to guarantee the portability. Table 2 lists the organizations that have provided spectra to date. Table 3 through Table 6 provides a listing of spectral data identified for ideal and non-ideal situations. Other international organizations are encouraged to provide additional spectra to expand and complete this collection.

Table 1 Structure of the depository for the test spectra.

Section 1 – Ideal spectra	
Section 1a – Uranium spectra	
Material types	Depleted uranium; e<0.7%, Natural uranium; e = 0.7% LEU; e = (0.7%, 2%), LEU; e = (2%, 5%), LEU; e = (5%, 20%) HEU; e = (20%, 35%), HEU; e = (35%, 60%), HEU; e = (60%, 90%) HEU; e > 90%, Reprocessed uranium
Detector types(1)	HPGe planar, HPGe extended range, HPGe coaxial, CZT, NaI, $LaBr_3$
Section 1b – Plutonium spectra	
Material types	Weapons grade; (r_{239}~94%, r_{240}~6%) Low burnup; (r_{239}~80%, r_{240}~19%) Medium burnup; (r_{239}~60%, r_{240}~25%) High burnup; (r_{239}~50%, r_{240}~27%) MOX; (Pu:U~1:40), MOX; (Pu:U~1:20) MOX; (Pu:U~1:10), MOX; (Pu:U~1:3)
Detector types(1)	HPGe planar, HPGe extended range, HPGe coaxial, CdTe, $LaBr_3$
Section 2 – Spectra acquired in non-ideal conditions	
Anomalies	Poor statistics, Poor resolution Too high count rate (dead-time, pile-up), Bad electronic setting Strong attenuators, Non-infinite thickness, High background
Section 3 – Unusual samples	
Material types	Pu from extremely high burnup Recycled MOX, Aged Pu with high Am Pure isotopes; (Pu_{239}>99.9%), Pure isotopes; (Pu_{240}>99.9%) Pure isotopes; (Am_{241}), Pure isotopes; (Pu_{238}>80%) Freshly separated U, Freshly separated Pu Contamination from FP, Contamination from MA Inhomogeneous U waste, Inhomogeneous Pu waste Inhomogeneous U/Pu waste

(1) Ideally a spectrum for any combination detector/material type.

4. Recommendations

Beyond accumulating an agreed upon set of spectra that can be used to evaluate the performance of the isotopics codes, producing a standard test method and a best practice guide are the next key steps to proceed forward. In addition, it is important to note that the upkeep and maintenance of these codes requires funding. The codes that are commonly used by the international community were developed within the framework of the U. S. national labs. Codes were initially developed in the early 1970s and continued through the early 1990s. Funding for continued development was decreased as the codes were transferred to commercial vendors. Funding should be made available to develop the scope of the next set of modifications that are necessary to meet the demands of the advanced nuclear fuel cycle. Federal funding is also needed to transfer and sustain the technical knowledge to the next generation of spectroscopy experts. Commercial vendors do not have access to the materials and facilities needed to advance these codes. This will ensure that the capability of these codes is advanced consistent with the measurement need. Spectroscopy experts at the research institutions should coordinate advancements to the codes with the international working group. This will ensure that codes are developed in a quality manner in accordance with standard test methods prior to release to the end user. The international working group should be sustained to provide a mechanism for coordinating interaction between the code developer and other end users.

Table 2: Legend of Spectra Suppliers

LEGEND					
US		**EC**		**South America (SA)**	
LLNL	A	Berlizov	B	ABACC	A
Sampson	B	ESARDA-DB	E	LASAL	L
Gunnink	C	JRC	J		
ORNL	D				
LANL	E				
Canberra	F				
Aquila	G				

Table 3: Unusual Samples Identified for IWG-GST Evaluation

ID	Description			
	Unusual isotopics			
		US	EC	SA
VHIBURNP	Extremely high burnup Pu			
RECYLMOX	Recycled MOX			
HIAMERPU	Aged Pu with high Am	B		
	Pure isotopes			
PURPU239	Pu-239>99.9%	F	E	
PURPU240	Pu-240>99.9%	F	E	
VHIAM241	Am-241	E, F		
HEATSRCE	Pu-238>80%	F		
	Freshly separated			
FRESHURA	U not in secular equilibrium		E	L
FRESHPLU	Freshly separated Pu			
	Contaminants			
FISSPROD	Presence of fission products			
MINORACT	Presence of minor actinides	E		
	Inhomogeneous materials (f.i. wastes)			
INHGURWA	Inhomogeneous U waste samples			A
INHGPUWA	Inhomogeneous Pu waste samples			
MIXUPUWA	Mixed U and Pu waste samples			

Table 4: Ideal Uranium Spectra Identified for Evaluation by IWG-GST

ID	Enrich Range (wt. %)	HPGe-Planar			HPGe-BEGe		HPGe-coax		CZT			NaI			LaBr3		
		US	EC	SA	US	EC	US	EC	US	EC	SA	US	EC	SA	US	EC	SA
DEPLURAN	0 to 0.7	B,D, E, F	E, B	L, A	F		D, E, F	E				D, F		L	D, F		L
NATLURAN	0,71	B,D, E, F	E, B	L, A	F		D, E, F	E			A	D, F		L, A	D, F		L, A
LEU00702	0.7 to 2	B,D, E, F	E, B, J	L, A	F		D, E, F	E			A	D, F		L, A	D, F		L, A
LEU02_50	2 to 5	B,D, E, F	E, B, J	L, A	F		D, E, F	E			A	D, F		L	D, F		L
LEU05_20	5 to 20	B, E, F	E, B, J	L			E	E									
HEU20_35	20 to 35	B,D, E, F	J				D, E					D, F			D, F		
HEU35_60	35 to 60	B,D, E, F	B, J	L			D, E					D, F			D, F		
HEU60_90	60 to 90	B, E, F		L			E										
HEUGRT90	>90	B, D, E	E, B, J	L			D, E	E				D, F			D, F		
REPRURAN	All																
REPEATCS	Any	B, F										F			F		

Table 5: Ideal Plutonium Spectra Identified for Evaluation by IWG-GST

ID	Pu239/Pu240 or Pu:U	HPGe-Planar		HPGe-coax		BEGE		CdTe		LaBr3	
		US	EC	US	EC	US	EC	US	EC	US	EC
WEAPGRAD	94/6	A,B, F	E, J	A,B, F	E, J	A,B, F		A		A	
LOBURNUP	80/19	A,B, F	E, J	A,B, F	E, J	B, F		A		A	
MEBURNUP	60/27	A,B, F	E, J	A,B, F	E, J	B, F		A		A	
HIBURNUP	50/x	B		B							
MOX01_40	1:40	B, F		B							
MOX01_20	1:20	B, F	E, J	B, F	E	F					
MOX01_10	1:10	B, F	J	B							
MOX01_03	1:3	B, F	J	B							
REPTPUCS	Any	F		F		F					

Table 6: Non-ideal Spectra Identified for Evaluation by the IWG-GST

ID	Description	Detector Type																	
		COAX			Planar			BEGE			CZT			NAI			LABR		
		US	EC	SA	US	EC	SA	US	EC	SA	US	EC	SA	US	EC	SA	US	EC	SA
POORSTAT	Poor statistic	D, E			E, F			F											
POORRESL	Poor resolution	D, E, F			E, F			F									F		
HIDEADTM	Too high count rates (high dead-time, pile up)	B, E			E														
POORSHAP	Bad electronic setting	E			E, F														
HIATTENS	Strong attenuators	B,D			D, F	B	A	F						D, F		L	D, F		L
NONINFTK	Non-infinite thickness					B	L												
HIBKGRND	High background															A			

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