

### Abstract

As part of regular laboratory operations, GBL15 (the UK CTBTO certified radionuclide laboratory) participates in annual Proficiency Test Exercises (PTEs) organised by the CTBTO. GBL15 consistently achieves the top 'A' grade in these exercises using dedicated HPGe detector systems; to achieve the sensitivity required these are specifically designed to use ultra-low background materials, and are further enclosed within advanced passive and active shielding [1].

GBL15 also houses a research system that consists of two high-efficiency HPGe detectors collocated in a single shield, which is currently capable of measuring and quantifying X-ray-gamma  $(X-\gamma)$  and gamma-gamma  $(\gamma-\gamma)$  signals [2-4]. By measuring cascades of gamma radiation, the background is dramatically lowered, greatly increasing the sensitivity of the system. This poster describes the use of this system to conduct a PTE, and compares the results to a standard analysis.

#### System

The  $\gamma$ - $\gamma$  coincidence system consists of two large area, planar HPGe detectors (model BEGe-6530, from MIRION - formerly Canberra UK - in Harwell, Oxfordshire) situated in a face-to-face configuration (both surrounding a central source), and enclosed within a 150 mm low-background Pb cave to minimise terrestrial radiation. Each detector signal is passed to two separate acquisition systems for redundancy and testing purposes; a DT5724D Desktop Digitiser (CAEN S.p.A., Italy), and two LYNX<sup>™</sup> units from Canberra.

The entire system is also covered in four 55 cm x 55 cm Scionix BC408 plastic scintillation plates connected to a Scionix preamplifier junction box. All four plates are controlled by another Canberra Lynx MCA, which generates an ICR logic pulse that is fed into the DT5724D Desktop Digitiser and the two LYNX<sup>™</sup> units. These act as a Cosmic Veto, removing events in the HPGe detectors that have a cosmic origin. This system typically reduces the background by 60-80% [5,6].



A rack and pinion system allows the lid to be interlocked over the detectors. Custom 3D printed sample holders are used to suspend the samples between the two detectors, creating a reliable and well characterised geometry. Both detectors are electrically cooled.

# **Control and Optimisation**

Previously, the system was configured experimentally, manually varying the delay and gate settings to optimise the coincidence window. Whilst effective, this was a time consuming and somewhat tedious task. Fortunately, GBL15 support to CTBTO (see poster T3.1-P27 for more details) recently produced a tool that is both easy to use and highly effective. This custom control and optimisation software was developed using the Python libraries provided by the Canberra Lynx Software Development Kit (SDK). The software can:

- Provide easy access to the Canberra MCA, including control of all acquisition functions

- Automatically optimise the delay and gate functions for coincidence systems
- Backup spectra at regular intervals, writing CNF's or PHD's with all sample information
- Take control of multiple LYNX<sup>™</sup> units, synchronise their internal clocks and perform list-mode acquisition



# Setup of timing for coincidence events

- the  $\gamma$ - $\gamma$  system:
- and that they were properly synchronised



# γ-γ Coincidence Analysis of the 2015 PTE at GBL15

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# **Processing the Data**

Two rounds of setup were performed for

1) The software was used to configure the delay and gate for each LYNX™ unit connected to the Cosmic Veto

2) The software was then used to confirm that  $\gamma$ - $\gamma$  coincidence spectra could be obtained from the system,

List-mode data acquisition allows a complete replay of the entire measurement, event-byevent. It also creates incredibly large files which require specialised software to process. The (unimaginatively named) 'Analysis Package' was developed by GBL15 for this purpose, and provides an all-in-one suite for the processing and analysis of complex list-mode data.



# From Peak Counts to Nuclide Activities

The final piece of the puzzle requires the transformation of a peak area ( $N_{peak}$ ) into a robust, reliable activity measurement. For routine single detector analysis, the factors required for this transformation are well known; b (branching ratio/abundance), t (acquisition time),  $\epsilon$  (peak efficiency), and c (correction factors to account for cascade summing and nuclide decay). In such a simple case, the activity, *A*, is calculated as:

$$A = \frac{N_{peak}}{b\varepsilon ct}$$

The activity of a two detector system (in singles mode, with a gain matched sum spectrum) is calculated in the same way, however the peak areas and efficiencies are also summed. This has the advantage of improving counting statistics, and reducing the impact of potential inhomogeneity in the sample. The downside is that you have twice the background, which limits the MDA improvement possible.

Using a coincidence pair, the background can be reduced by many orders of magnitude. The difficulty lies in calculating the appropriate factors to quantify such a signal. RIMMER (Randomised Iterative Monte-Carlo Model of ENSDF Records) was developed specifically for this [2,3,4].

#### Running RIMMER

- The factors were calculated using **RIMMER** in batch mode
- Firstly the peak and total efficiencies were generated using ISOCS, a Canberra product for characterised detectors
- A library file was then supplied to RIMMER along with the efficiency data
- A convenient csv format file was generated with all the factors required to quantify all possible emissions from the nuclide library for both singles and coincidence analysis

orary Parameters: 1inimum Energy (keV) = 10.0 1inimum Cascade Fraction = 1.0E-06 E: Add Efficiency P: Plot Efficiency er your choice: \_

RIMMER is scalable to any multiplicity of detector system, and has also found use generating Cascade Summing Factors (CSFs) at GBL15, with a library of over 250 nuclides calculated for every sample geometry and detector combination in the laboratory [8].

- The Analysis Package sorts the listmode data into a number of linked TTrees (complex ntuples in ROOT [7]), greatly reducing the disk space required
- Histograms are produced for each detector in standard, coincidence and anti-coincidence modes
- Standard, coincidence, and anticoincidence spectra are also created for a fully calibrated and (software) gain matched sum of all detectors
- Coincidence matrices are produced, and used to extract coincidence events from user-supplied energy dates
- Complex peak fitting is also available using a number of functions, including Gaussian, Voigt, and Lorentzian. All can include exponential tails



Once the PTE had been counted on both certified CTBTO detectors at GBL15, it was also measured in list-mode on the  $\gamma$ - $\gamma$  system. Analysis was carried out independently, with different analysts completing the standard analysis pipeline and the coincidence system analysis pipeline respectively.

Table 1 shows the results obtained using a CTBTO certified laboratory system (Canberra BEGe-5030 detector, ultra-low background shielding, ~4 $\pi$  Cosmic Veto), the  $\gamma$ - $\gamma$  system in 'super-singles' mode (software gain-matched and calibrated), and the  $\gamma$ - $\gamma$  system in coincidence mode. Note that some nuclides could not be obtained in coincidence mode due to the lack of a coincidence signature (this is the nuclides fault, not ours).

				GBL15 Activity Results (ratio to reference value)							
Nuclide	Classification	Reference Activity		GBL15_002		Dual Gamma (singles)		Dual Gamma (coincidence)			
		Activity (Bq)	Unc (%)	Activity	Unc (%)	Activity	Unc (%)	Activity	Unc (%)		
Zn-65	Major	1.87E+02	1.4	0.97	3.1	1.03	3.60	-	-		
Nb-95	Major	2.72E+01	8.88	0.94	3.7	0.99	3.04	-	-		
Zr-95	Major	7.71E+01	2.08	0.95	3.3	0.99	3.14	-	-		
Ru-103	Major	1.05E+03	2.16	0.96	4.4	0.94	3.14	0.98	0.05		
Ru-106	Major	1.13E+02	5.92	0.94	6.8	0.96	3.56	1.01	0.04		
Sb-124	Major	1.01E+01	3.7	0.96	5.4	1.01	4.50	1.01	0.04		
Sb-125	Major	1.59E+01	4.66	0.90	6.6	0.97	3.03	0.99	0.06		
Cs-136	Major	3.35E+02	2.42	0.97	10.7	1.03	3.86	0.99	0.05		
Cs-137	Major	6.53E+00	4.56	1.00	3.6	1.00	4.35	-	-		
Ba-140	Major	5.04E+02	2.16	0.96	4.3	1.00	4.67	1.01	0.07		
La-140	Major	5.81E+02	2.08	0.96	3.3	1.00	3.32	1.03	0.05		
Ce-141	Major	2.30E+02	2	0.97	4.7	1.02	4.88	-	-		
Ce-144	Major	1.35E+01	3.05	0.95	5.8	1.02	4.91	-	-		
Nd-147	Major	6.00E+01	10.07	0.95	7.9	1.04	3.72	1.01	0.07		
Ag-111	Minor	2.87E+02	2.68	0.98	5.8	0.96	3.30	0.99	0.07		
Sn-125	Minor	1.60E+02	34.02	0.93	36.0	1.00	35.00	1.02	30.05		
Sb-126	Minor	1.00E+02	3.9	1.02	12.2	0.97	14.30	0.95	8.05		
Te-129	Minor	2.37E+02	10.07	0.87	9.1	1.08	8.49	1.04	7.07		
Te-129m	Minor	3.77E+02	10.07	1.00	57.3	1.01	53.79	1.05	50.05		

In Table 1, results shown in green are within 5% of the (corrected) reference value. Those highlighted in yellow show those results that fall outside of this but within 10%. All results pass the % difference, Zeta and Uncertainty ratio tests. Some errors appear artificially large; this is often due to errors quoted in the nuclear data which are propagated through to the activity measurement. Overall, the  $\gamma$ - $\gamma$  system performed as least as well as a conventional system, and allowed far greater confidence when confirming the presence of radionuclide signatures. Table 2 (below) shows the grade achieved in the 2015 PTE – an A grade would have also been achieved with the  $\gamma$ - $\gamma$  system.

Participant	%Difference (No. of Passing)	Zeta score (No. of Passing)	False negative (No. of missed nuclides)	False Positive (No. of False Positive)	RL test (No. of outliers)	Overall grade
ARL01	Fail (9)	Fail (9)	Fail (1)	Pass (0)	Pass (0)	F
AUL02	Pass (13)	Pass (14)	Pass (0)	Pass (0)	Pass (0)	А
ATL03	Pass (13)	Pass (13)	Pass (0)	Pass (0)	Pass (0)	А
BRL04	Pass (14)	Pass (14)	Pass (0)	Pass (0)	Pass (0)	А
CAL05	Pass (12)	Fail (10)	Pass (0)	Pass (0)	Pass (0)	В
CNL06	Pass (12)	Pass (13)	Fail (1)	Pass (1)	Pass (0)	В
FIL07	Pass (11)	Pass (12)	Pass (0)	Fail (2)	Pass (0)	A-
FRL08	Pass (14)	Pass (14)	Pass (0)	Pass (0)	Pass (0)	А
ILL09	Pass (13)	Pass (13)	Pass (0)	Pass (1)	Pass (0)	А
ITL10	Pass (14)	Pass (14)	Pass (0)	Pass (0)	Pass (0)	А
JPL11	Pass (14)	Pass (13)	Pass (0)	Pass (0)	Pass (0)	А
NZL12	Pass (12)	Pass (12)	Fail (1)	Pass (1)	Fail (1)	В
RUL13	Fail (11)	Pass (14)	Pass (0)	Pass (0)	Fail (1)	A-
ZAL14	Fail* (0)	Fail* (0)	Pass (0)	Pass (0)	Fail (2)	С
GBL15	Pass (14)	Pass (14)	Pass (0)	Pass (1)	Pass (0)	А
USL16	Pass (13)	Pass (14)	Pass (0)	Pass (0)	Pass (0)	A

For the first time, a PTE sample has been measured and analysed using a  $\gamma$ - $\gamma$  system. The results demonstrate that quantification is reliable and robust in both singles and coincidence modes. The availability of both modes gives the user much greater confidence in nuclide identification. Coincidence signals are also much 'cleaner' than those in a singles spectra due to the greatly reduced background.

A full measurement and analysis pipeline has also been created and tested. Using nothing but raw nuclear data in the form of ENSDF records and a detector system, GBL15 can now carry out the complete cycle of measurement, data processing and spectral/matrix analysis for an arbitrarily complex system.

The ability to identify and quantify radionuclide signatures via two different methods simultaneously is completely unique, and will have a number of interesting applications. One such application is a 'measurement restriction' system, where absolute confidence in the result is crucial. We hope to demonstrate such a system based upon these combined techniques soon.

For more information on any of our research/software, please contact the authors.

[1] Burnett J.L, Davies A.V, (2013) J Radioanal Nucl Chem 298-2, 987-992 [2] Britton R, Burnett J.L, Davies A.V, Jackson M, (2015) J Environ Radioact 146, 1-5 [3] Britton R, Jackson M, Davies A.V, (2015) J Environ Radioact 149, 158-163 [4] Britton R, Jackson M, Davies A.V, (2016) J App Rad Isot 116, 128–133

[5] Science and Technology Conference 2017, Poster – T3.1-P27 [6] Burnett J.L, Davies A.V, (2014) Nucl Instrum Methods Phys Res A 747, 37-40 [7] Brun R, Rademakers F, (1997) Nucl. Instrum. Methods Phys Res A 389, 81-86

# BD SCIENCE AND TECHNOLOGY CONFERENCE

# Measurement and Analysis of PTE2015

# Conclusion

<sup>[8]</sup> Jackson M, Britton R, Davies A.V, McLarty J.L, Goodwin M, (2016) Nucl Instrum Methods Phys Res A 834, 158-163