High resolution $\beta - \gamma$ coincidence spectrometry at the UK CTBT Radionuclide Laboratory

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Abstract. Detection of radioxenon is often considered the most probable indicator of an underground nuclear explosion. GBL15 is the UK’s Comprehensive Nuclear-Test-Ban Treaty Certified Radionuclide Laboratory, operated at AWE Aldermaston and has a history of developing high fidelity coincidence detection systems for particulate radionuclides. The Laboratory also operates a SAUNA II system, using NaI(Tl) and plastic scintillator detectors to measure $\beta - \gamma$ coincidences from the decay of the four radioxenon isotopes, namely $^{133}$Xe, $^{135}$Xe, $^{131m}$Xe & $^{133m}$Xe. Here the efforts to date in exploring new technologies for next generation laboratory-based $\beta - \gamma$ coincidence spectrometry for radioxenon measurements are discussed. Results are presented from preliminary measurements using a PIPSBox detector with a high purity germanium $\gamma$ detector and the output compared to that of lower resolution systems. This investigation will be used to scope future programmes on the technology used for the measurement of radioactive noble gas nuclides at GBL15.

1. Introduction
Beta-delayed $\gamma$-ray coincidence measurements have become the go-to technique for detection and quantification of radioxenon isotopes as a signature of nuclear weapons testing [1]. Since 1999, the International Noble Gas Experiment (INGE) has pioneered the development of systems for the collection, quantification and measurement of environmental radioxenon gas samples, and to date there are 25 certified systems incorporated in to the International Monitoring System (IMS) - a global network of detectors designed to detect a nuclear explosion in breach of the Comprehensive Nuclear-Test-Ban Treaty (CTBT) [2, 3]. As one of the 16 IMS Radionuclide Laboratories, GBL15 (the UK CTBT Certified Laboratory) is tasked to measure both particulate and noble gas samples collected on the IMS. This is currently achieved using high purity germanium (HPGe) detectors for particulate filter measurements, and a Swedish Automatic Unit for Noble Gas Acquisition (SAUNA) II system for radioxenon measurement [4].

The radioxenon isotopes (and long-lived isomers, herewith also referred to as ‘isotopes’) relevant to nuclear weapons testing on the IMS are $^{133}$Xe, $^{135}$Xe, $^{131m}$Xe & $^{133m}$Xe [5]. Figure 1 shows the distribution of fission product yield, by baryonic mass number, $A$. Radioxenon isotopes are in a crucial region, on the crest of the cumulative fission yield curve; and if measured soon enough after the nuclear event zero time, they can be used to discern between the nuclear fissile materials used. There are other properties, both chemical and physical that make these...
Figure 1. Comparison of cumulative (neutron-induced) fission product yields for each atomic mass, from selected nuclear fissile materials, with different neutron energies (T: Thermal, H: 14 MeV). Radioxenon isotopes are produced with relatively high yields, making them an ideal nuclear weapons test signature. Figure produced using data from [6].

radionuclides ideal signatures for nuclear weapons test monitoring; the low chemical reactivity of xenon greatly increases the likelihood of atoms escaping from an underground nuclear test (UGT) cavity when compared to particulate matter. The nuclear half-life ranges from 9 hours to 11 days for the four relevant isotopes, meaning detections could take place many weeks after the event. All 4 radionuclides have a selection of radiative emissions including X/$\gamma$-ray and $\beta^-/e^-$ which are all measurable and distinguishable using today’s radiation counting equipment.

The SAUNA II detector system used at GBL15, incorporates a BC404 plastic scintillator gas cell within a NaI(Tl) crystal to perform near 4$\pi \beta - \gamma$ measurements. Whilst the plastic scintillator $\beta^-/e^-$ detector provides just enough $e^-$ energy resolution to resolve the 129 keV and 199 keV internal conversion electrons from the metastable radioxenon isotopes $^{131m}Xe$ & $^{133m}Xe$, the interferences associated with lower resolution systems have driven the need for a high-resolution solution. This applies also to the NaI(Tl) detector, which is capable of measuring the low energy X-ray region (29-35 keV) with high efficiency, but is unable to resolve individual contributions from Xe and Cs daughters. This approach to radioxenon measurement has been taken by a number of groups and a new higher resolution noble gas measurement system (currently undergoing testing) will soon be incorporated into the IMS.

2. $\beta - \gamma$ comparison

Low-resolution spectrometry usually offers a number of benefits including cost, ruggedness and detection efficiency. Increasing detection efficiency involves more than selecting the most efficient detector, one must consider the unresolved interferences between emissions which are otherwise resolved using higher resolution systems. It is often useful to combine emissions from radionuclides in order to enhance the signal. These should be well-resolved emissions, originating solely from the target radionuclide, else the background signal is also increased, having a negative effect on the overall detection limit. X-ray emissions are element specific and, at these resolutions, not isotopically selective; for example $^{133}Xe(Cs)$ X-rays interfere with the measurement of metastable isotopes $^{131m}Xe$ & $^{133m}Xe$ which are quantified using the Xe X-rays
Figure 2. Left: (upper) Raw and total coincidence $\gamma$ spectrum for the measurement of a mixed isotope radioxenon sample and (below) a $\beta$ spectrum showing raw, total coincidence and gated coincidence data (gated 20-40 keV). Right: $\beta - \gamma$ coincidence matrix of a mixed isotope radioxenon sample. Inset is a zoom on the X-ray (29-35 keV) and $^{133}$Xe region (81 keV). One can distinguish the discrete electron signatures in the $\beta$ axis, but the X-rays form a single, combined signal. Data collected using a SAUNA II system.

in the same region. The SAUNA system provides reasonable detection efficiency (around 70 % in the coincidence regions) but the presence of $^{133}$Xe in a mixed isotope radioxenon sample hinders the measurement of $^{131m}$Xe & $^{133m}$Xe.

2.1. Current Measurement Systems
The use of $\gamma$ singles spectrometry for radioxenon measurements is still taking place on the IMS, but all current developers are now looking to make use of $\beta - \gamma$ coincidence systems. The two main suppliers of current systems, Scienta (Sweden, ‘SAUNA’) and CEA (France, ‘SPALAX’) have developed next generation IMS systems which are currently under testing. The new CEA system uses a silicon wafer $\beta$ detector and broad energy germanium (BEGe) detector in coincidence and is the first high resolution $\beta - \gamma$ system to be used for CTBT monitoring. The silicon wafers are held inside a gas-tight cell with carbon fibre windows for minimal X/$\gamma$ attenuation, in a detector known as the PIPSBox.

2.2. PIPSBox
In 2013 scientists from CEA (France) published a paper describing a new electron detector, developed in conjunction with Canberra (Mirion Technologies), based on a gas cell containing two large passivated implanted planar silicon (PIPS) wafers [7]. The detector offered excellent electron resolution (3.3 % at 45 keV) and a good efficiency of detection. Increasing the electron resolution allows for better discrimination between the conversion electron (CE) emissions of $^{131m}$Xe and $^{133m}$Xe at 129 keV and 199 keV respectively, which leads to less interference and hence a lower detection limit.

The PIPSBox is designed with 500 $\mu$m silicon to maximise detection efficiency of $\beta^-$ particles whilst ensuring high conversion electron energy resolution. CEA demonstrated a PIPSBox–HPGe system where the coincidence window was set to 10 $\mu$s, which was able to capture a good
coincidence-signal to random-coincidences ratio, but did not give a reason for such a long time window. Some of the background coincidence regions had zero counts after 2 days of background measurement, leading to very low detection limits (around 0.002 Bq after 1 day) which is around 70 times greater than that which is possible using $\gamma$ singles. Quoted Minimum Detectable Concentrations (MDCs) for $^{131m}\text{Xe}$ and $^{133m}\text{Xe}$ are 0.018 mBq m$^{-3}$ based on a 1 day measurement, which is around a factor of 30 and 10 times improved from the previous SPALAX system. It has been noted that the $^{133}\text{Xe}$ 45 keV CE peak had lower resolution than expected and is likely due to higher predominance of energy straggling at low energy, following loss of energy of electrons in the gas mixture [8].

Other institutions have investigated the possibility of using silicon for $\beta$ measurement in coincidence with $\gamma$ in a variety of geometries [9, 10] where simulation showed that the probability of full energy deposition of an electron of energy 45 - 346 keV in silicon of thickness 450 $\mu$m, is 12.1 - 12.4 % (at $\sim$276 K). This is a considerable drop in efficiency of the $e^-/\beta^-\gamma$ detector when compared with plastic scintillators and could become the sensitivity-limiting factor. The benefits of high resolution $\beta-\gamma$ spectrometry for radioxenon measurements have been demonstrated by CEA [11]. GBL15 has procured a PIPSBox detector and here we present some of the results of a PIPSBox setup alongside a BEGe detector, specified for radioxenon measurements in the laboratory.

### 3. Quantifying radioxenon signatures

| Electron Signal | Electron Energy (keV) | Photon Signal | Photon Energy (keV) | Parent Isotope |
|----------------|----------------------|---------------|---------------------|----------------|
| $\beta^-$      | 915.3                | X             | 35.972              | $^{135}\text{Xe}$ (Cs) |
| $\beta^-$      | 915.3                | X             | 35.252              | $^{135}\text{Xe}$ (Cs) |
| $\beta^-$      | 346.4                | X             | 35.972              | $^{135}\text{Xe}$ (Cs) |
| $\beta^-$      | 346.4                | X             | 35.252              | $^{135}\text{Xe}$ (Cs) |
| C.E.           | 129.366              | X             | 33.562 - 34.552     | $^{131m}\text{Xe}$ |
| C.E.           | 198.655              | X             | 33.562 - 34.552     | $^{133m}\text{Xe}$ |
| $\beta^-$      | 915.3                | X             | 30.973              | $^{135}\text{Xe}$ (Cs) |
| $\beta^-$      | 915.3                | X             | 30.625              | $^{135}\text{Xe}$ (Cs) |
| $\beta^-$      | 346.4                | X             | 30.973              | $^{135}\text{Xe}$ (Cs) |
| $\beta^-$      | 346.4                | X             | 30.625              | $^{135}\text{Xe}$ (Cs) |
| C.E.           | 129.366              | X             | 29.459, 29.779      | $^{131m}\text{Xe}$ |
| C.E.           | 198.655              | X             | 29.459, 29.779      | $^{133m}\text{Xe}$ |
| $\beta^-$      | 346.4                | X-Ge          | 21.5, 26.5          | $^{133}\text{Xe}$ (Cs) |
| $\beta^-$      | 915.3                | X-Ge          | 21.5, 26.5          | $^{133}\text{Xe}$ (Cs) |
| C.E.           | 129.366              | X-Ge          | 20.0, 24.0          | $^{131m}\text{Xe}$ |
| C.E.           | 198.655              | X-Ge          | 20.0, 24.0          | $^{133m}\text{Xe}$ |

The SAUNA II system is one of the most efficient solutions for radioxenon detection and is currently in use at the GBL15 noble gas laboratory. The radiation measurement data is interpreted by selecting regions of interest (ROI) and summing the counts in each region.
3.1. The Net Count Correction Method

Within the Noble Gas Treaty Verification Community, the Net Count Correction Method (NCC) is used to quantify radioxenon activities from the $\beta - \gamma$ coincidence histogram. In order to do so, a number of corrections must be taken into consideration. The NCC method was developed specifically for this subject. Currently, the NCC method is in use at a number of international institutions, where it is used for the analysis for noble gas $\beta - \gamma$ spectra from the IMS. The method is based on calculating the corrected count rate in pre-calibrated ROI, then converted to activity through values derived from calibration procedures and nuclear data. The original interpretation of the NCC method is best explained in an FOI published report (see [15]). To implement the NCC on high resolution coincidence spectra, a rigorous calibration procedure must be implemented, involving the measurement of radioxenon reference standards.

4. Experimental Setup & Results

The current working high resolution $\beta - \gamma$ system at GBL15 uses an Ortec BEGe and PIPSBox, alongside the Mirion LYNX Multi-Channel Analyser (MCA) to process the pulses. High-gain preamplifiers were selected from CAEN, to amplify the PIPS signal. The separate MCAs are time synchronised and the data is collected and stored as time-stamped events, which are then post-processed using custom GBL15 software.

![Figure 3. $\beta - \gamma/e^+\gamma$ coincidence spectrum showing coincidences between the X-rays from $^{131}$Xe, $^{133}$Xe, $^{133}$Cs & $^{135}$Cs and the $\beta^-$ & $e^-$ particles from the decay of parent isotopes. A time window of 6 $\mu$s was used, comparable to the 10 $\mu$s used in the literature.](image)

The high resolution coincidence matrix in figure 3 highlights signals that were not previously revealed. By order of decreasing photon energy, the coincidence signals can be defined as such: $^{133}$Cs X-ray & $^{133}$Xe $\beta^-$, $^{135}$Cs X-ray & $^{135}$Xe $\beta^-$, $^{131}$Xe X-ray & $^{131m}$Xe $e^-$ (129 keV), $^{133}$Xe X-ray & $^{133m}$Xe $e^-$ (199 keV), $^{135}$Cs X-ray & $^{133}$Xe $\beta^-$, $^{135}$Cs X-ray & $^{135}$Xe $\beta^-$, $^{131}$Xe X-ray
& $^{131m}$Xe $e^-$ (129 keV), $^{133}$Xe X-ray & $^{133m}$Xe $e^-$ (199 keV). The 45 keV internal conversion $e^-$ can be seen clearly atop the $\beta^-$-X coincidence signals.

Quantifying $^{133}$Xe and $^{135}$Xe is straightforward using the 81 keV and 250 keV $\gamma$ (in coincidence with $\beta^-$) respectively. This work shows that $^{131m}$Xe and $^{133m}$Xe can be almost entirely resolved from one another (see $e^-\text{-X}$ coincidences in figure 3), which allows for a very low interference between the regions. Through further experiment, by injection of different combinations of the radioxenon isotopes of interest (and radon progenies), and validated by modelling, the $\beta-\gamma$ efficiency can be quantified and the interference factors derived, as well as suggested developments to the analysis method, such as combining and summing different signals. Through post-analysis of the data, it is possible to investigate the physical processes taking place giving rise to previously undescribed spectral features.

5. Conclusions

To date, a review of current technologies for a next generation laboratory-based high resolution $\beta-\gamma$ coincidence system has been carried out, a prototype system developed using PIPSBox technology, and initial measurements have been carried out. This work has been discussed and presented at a number of meetings to date and upcoming experiments have been scoped, to include measurement of other (more prompt) fission product gases such as radiokrypton, Monte Carlo modelling of the prototype system using GEANT4 (along with other configurations), determination of detection limits and measurement uncertainties and advanced $\gamma-\gamma$ measurements using $\beta$ gating. The PIPSBox–HPGe system has provided a useful starting place for the project, which has delivered the first high resolution $\beta-\gamma$ coincidence measurements at GBL15.

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