

## Measurement of gaseous fission products on an electron-photon coincidence detector

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ABSTRACT

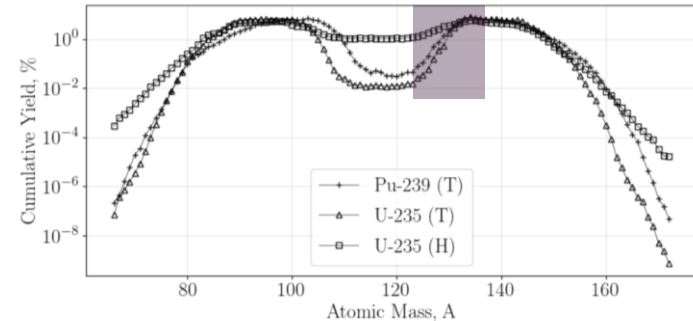
Gaseous fission products have been produced via neutron irradiation of a uranium target and extracted using a custom gas processing system for measurement on a prototype high-resolution beta-gamma coincidence detection system. The gas was extracted and measured in two stages in order to measure the prompt and delayed fission products.

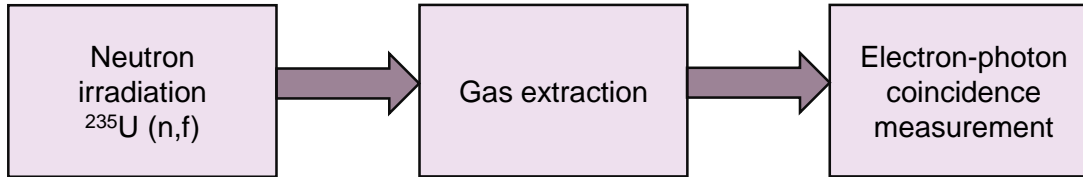
We present an overview of the system used to extract gaseous products, and the results of the advanced coincidence techniques used to identify and quantify the radionuclides present. This work demonstrates the capability to produce gaseous radionuclides for quality assurance and calibration purposes in Radionuclide Laboratories supporting the Comprehensive Nuclear-Test-Ban Treaty (CTBT) as well as for the calibration of equipment.

Fig. 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 from [1]. Data from England and Rider.

**Why?**

- Radioactive gases produced from the fission of <sup>235</sup>U can be indicative of a violation to the CTBT
- High-resolution  $\beta$ - $\gamma$  coincidence measurement systems can improve our measurement accuracy, to better understand the  $e^-$ -X and  $\beta$ - $\gamma$  coincidence signatures from the nuclear decay of radioxenon isotopes <sup>133</sup>Xe, <sup>135</sup>Xe, <sup>133m</sup>Xe and <sup>131m</sup>Xe
- The capability to produce, extract and measure gas samples is required for detector calibration and measurement traceability





**Summary**

- <sup>235</sup>U target irradiated with thermal energy neutrons at the NPL neutron facility
- Gas extracted at two points in time (red lines) at t=22 hours and t=70 hours
- High-resolution β-γ coincidence measurements conducted using PIPSBox-HPGe detector system
- Detected radioxenon isotopes <sup>133</sup>Xe, <sup>135</sup>Xe, <sup>133m</sup>Xe, <sup>135m</sup>Xe
- Detected radiokrypton isotopes: <sup>88</sup>Kr, <sup>85m</sup>Kr, <sup>85</sup>Kr
- Compare measured activity ratios with simulated system

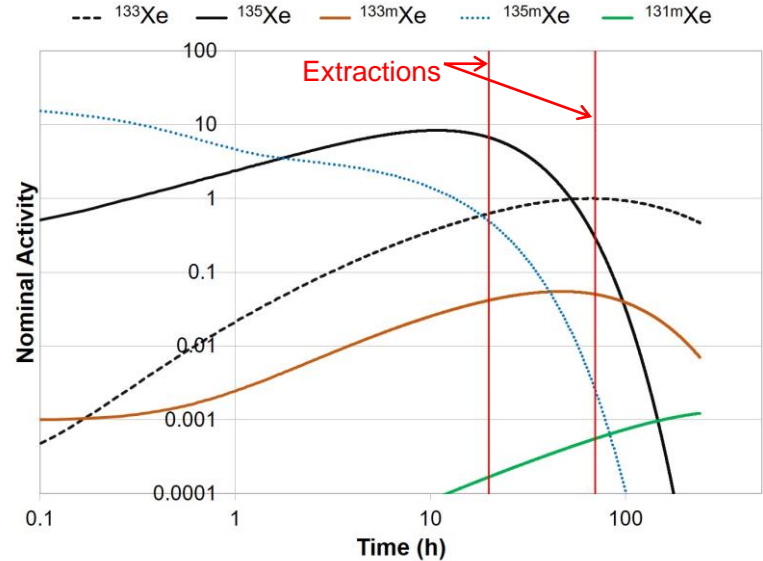


Fig. 2. (Right) Simulated activity of five radioxenon isotopes relative to <sup>133</sup>Xe maximum activity, using <sup>235</sup>U fission yields (with thermal energy neutrons) for 10 days. The vertical red lines denote the two points in time where extractions of gas were made. The activities here represent those in the target (i.e. unfractionated from parent nuclei).

Thermal neutrons (up to  $2 \times 10^7 \text{ n cm}^{-2} \text{ s}^{-1}$ ) are produced by accelerating  $^2\text{H}$  ions into Be targets located within a large graphite block. A gas-tight assembly loaded with the highly-enriched uranium target is placed within a narrow cavity inside the graphite block.

Following 3.0 hours of irradiation, the assembly is removed from the cavity and transferred to the counting laboratory. The fission product gases generated during irradiation are extracted from the assembly and combined with stable xenon (the 'extraction' phase). Volatile fission products are not extracted and mostly remain in the assembly. The extracted fission product gas is transferred by pressure-drop to the PIPSBox detector cell where it is sealed, and the acquisition is started.

LYNX Digital Signal Analysers (DSA) used for each detector

Time-stamped list mode acquisitions

Data sorted in C++/ROOT & extracted in Python

Following the acquisition, a file for each detector records each time-stamped count. The data is sorted and post-processed to generate the relevant coincidence and gated-coincidence spectra

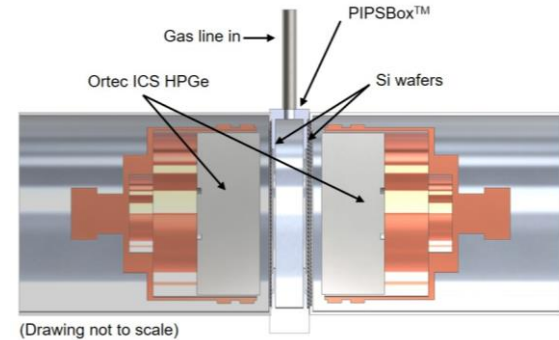
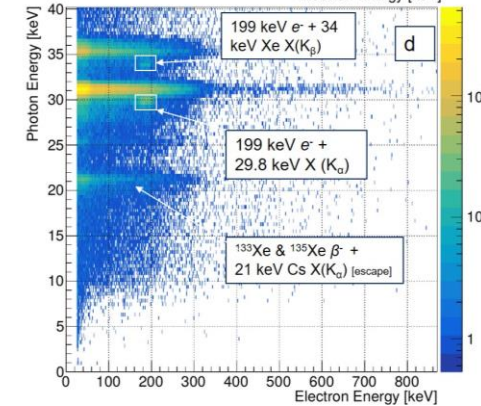
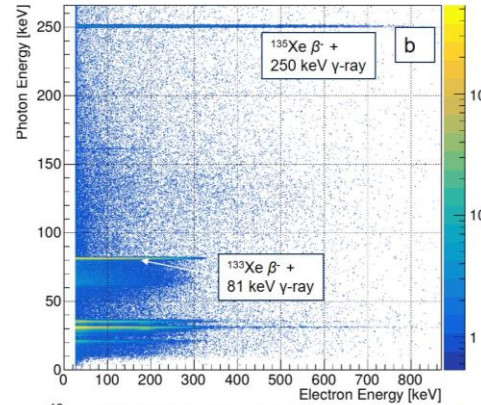
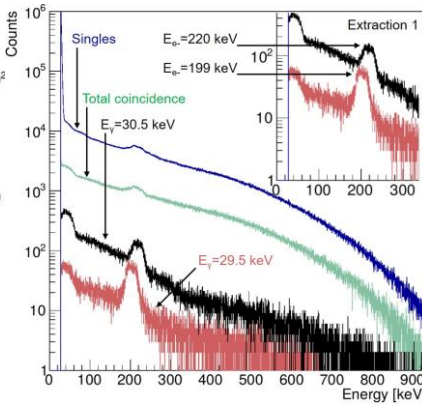
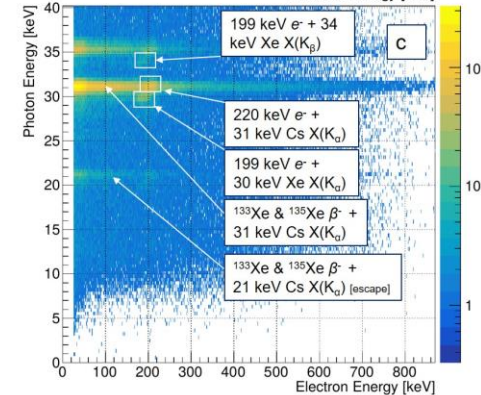
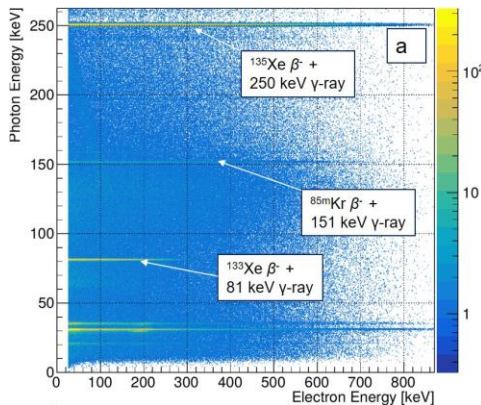


Fig. 3. Schematic diagram of the HPGe+PIPSBox setup used at NPL to measure the fission gas. The system runs with four detector channels; two photon channels and two electron channels.

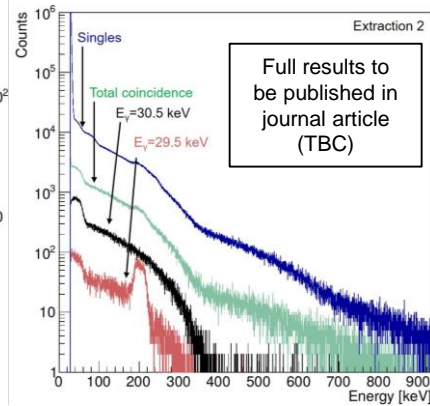
# RESULTS

**Blue:** singles e<sup>-</sup>-spectrum, **green:** total e<sup>-</sup>-coincidence (γ-gated) spectrum, **black:** 30.5 keV gate (Cs X<sub>K</sub>), **red:** 29.5 keV gate (Xe X<sub>K</sub>). The extraction 1 X(Cs) gate (**black**) highlights the 45 keV <sup>133</sup>Xe conversion electron, the 220 keV <sup>135</sup>Xe conversion electron and <sup>135</sup>Xe+<sup>133</sup>Xe β<sup>-</sup>-continuum (max. 900 keV). Conversion electron peaks are highlighted in the inset spectrum.



Electron-photon coincidence matrices for extraction 1 (a) and extraction 2 (b). Plots c and d are zoomed to the X-ray region. Identified coincidence signatures are labelled

The X(Cs) gate in extraction 2 shows only the <sup>133</sup>Xe β<sup>-</sup>-continuum (max. 350 keV). The X(Xe) gate (red) highlights the 199 keV C.E. from <sup>133m</sup>Xe in both spectra.



- Gaseous fission products have been produced via neutron induced fission of  $^{235}\text{U}(n,f)$  and extracted to a high-resolution  $\beta - \gamma$  coincidence spectrometry system for acquisition.
- Analysis of the spectra confirms the presence of the radioxenon isotopes of interest:  $^{133}\text{Xe}$ ,  $^{135}\text{Xe}$ ,  $^{133m}\text{Xe}$  as well as other noble gas radionuclides  $^{85m}\text{Kr}$ ,  $^{85}\text{Kr}$  and  $^{88}\text{Kr}$ .
- Features in the projected electron spectra have been identified, including internal conversion electrons from  $^{133m}\text{Xe}$  and  $^{135}\text{Xe}$ . The calculated activity ratio of  $^{135}\text{Xe}/^{133}\text{Xe}$  is similar to the expected values from simulations using fission yield data, allowing for differences due to ingrowth within the measurement cell, after fractionation.
- This work demonstrates the capability to produce noble gas fission products at NPL, which can be used to improve radioxenon measurement systems such as those used at AWE and in these measurements. This work will continue to support the development of the UK CTBT Radionuclide Laboratory capability, as well as support the development of the production of a  $^{133}\text{Xe}$  primary standard. This work is part of efforts by NPL and AWE to establish traceability for IMS radioxenon measurements.
- **See talk: O3.2-482 “A high-resolution laboratory-based  $\beta$ - $\gamma$  coincidence spectrometry system for radioxenon measurement”**

