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Radioactive gas metrology at NPL and the development of short-lived gas standards

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Radioactive gas metrology

NPL maintain the UK's primary standards of radioactivity; including radioactive gas standards. Gas standards of ^{3}H, ^{11}C, ^{14}C, ^{85}Kr and ^{133}Xe are realised by means of absolute counting using a series of length-compensated internal-gas proportional counters (see fig. 1). These standards are used for the calibration of stack, area and environmental monitors. Presented is an overview of recent work at NPL to produce and standardise the short-lived gases ^{41}Ar, ^{85m}Kr, ^{87}Kr, ^{88}Kr and ^{135}Xe for the calibration of a Fukushima NPP criticality monitor.

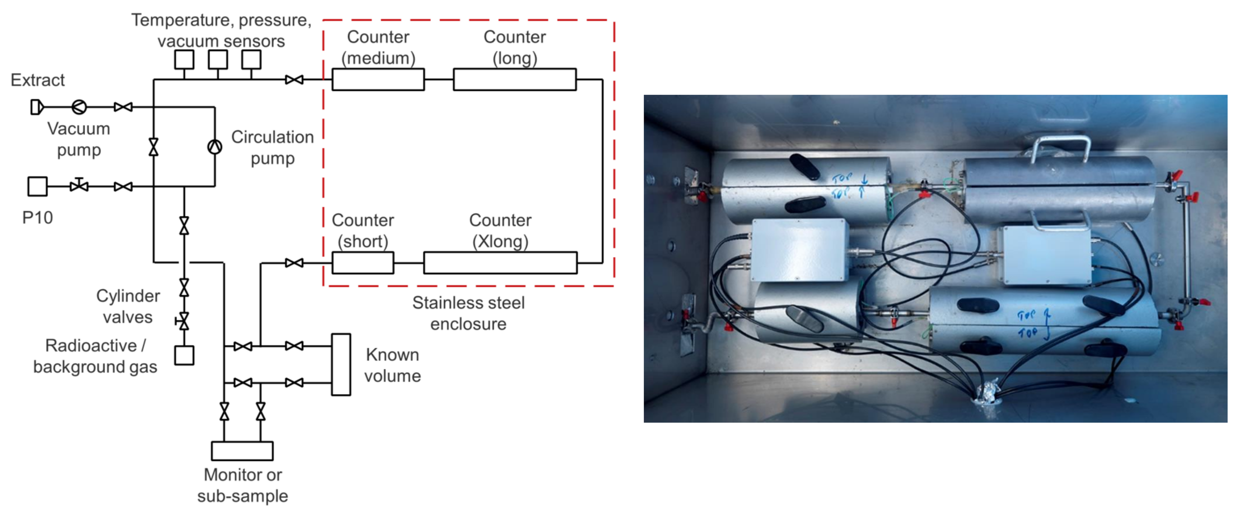

Fig. 1 – NPL absolute gas counting system.

Criticality monitoring at Fukushima

The Fukushima NPP reactors are in varying states of disrepair. During recovery, the sub-criticality of the debris must be monitored. Hitachi and Hitachi-GE are responsible for developing a criticality monitoring system for this purpose. As neutron detectors cannot monitor all locations within the PCV, a HPGe detector located in the turbine hall is used to monitor the fission product (FP) and activation product (AP) gases constantly flushed by N_2 from the reactor (see fig. 2).

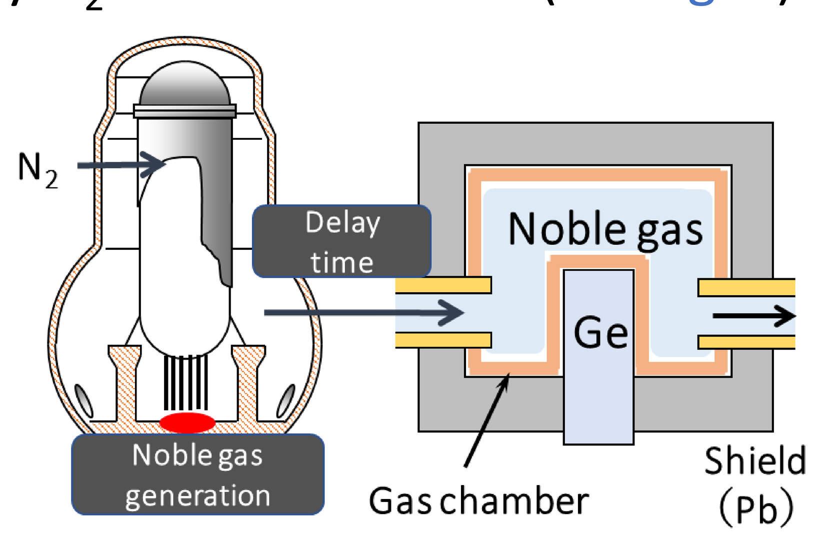

Fig. 2 – Noble gas monitoring system installed at Fukushima

Of most interest is the ratio of ^{88}Kr to ^{135}Xe. The high ^{134,137}Cs background in the turbine hall results in a low peak-to-background ratio for photopeaks below 800 keV. This make it difficult to detect criticality in short periods. Hitachi have requested NPL to produce a traceable standard of ^{88}Kr and to use this to calibrate the Hitachi monitor thus enabling quantification using the high energy photopeak at 2,392 MeV.

Production of noble gases at NPL

Short-lived FP and AP gas standards are produced at NPL by irradiating enriched ^{235}U or stable isotope precursors with thermal neutrons using the NPL Thermal Pile (see fig. 3a). Thermal neutrons (up to 2 × 10^{7} n cm^{-2} s^{-1}) are produced by accelerating deuterium ions into beryllium targets located within a large graphite block. The gas tight assembly (see fig. 3b), is placed within a narrow cavity inside the graphite block.

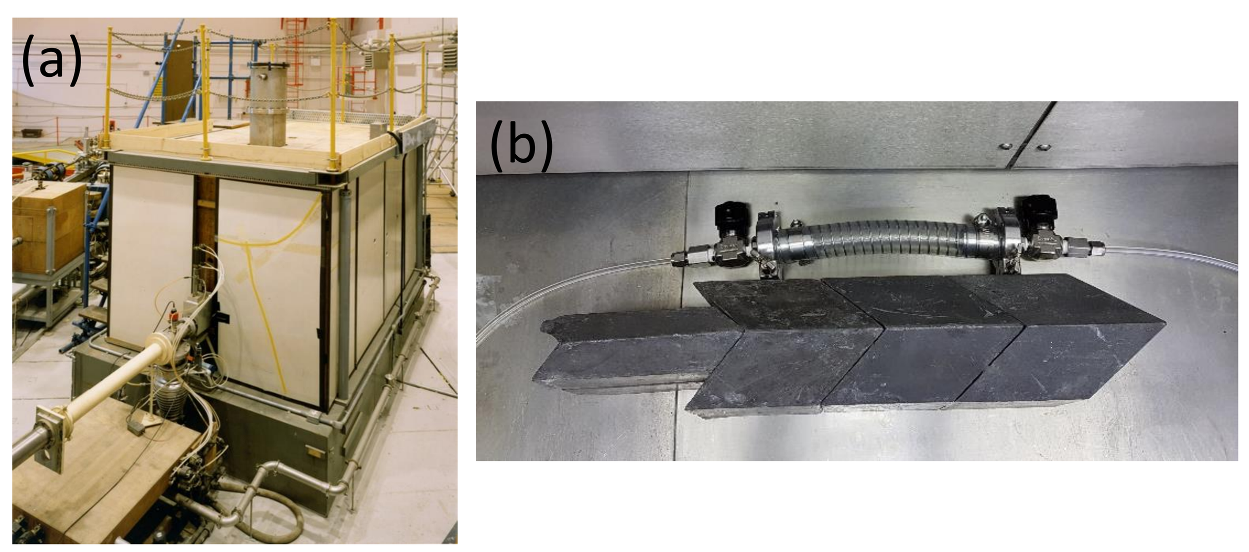

Fig. 3 – (a) NPL Thermal Pile and (b) ^{235}U target assembly.

Following irradiation, the FP or AP noble gases are extracted from the target assembly and transferred to the NPL absolute gas counting system for standardisation. An HPGe detector has been installed in-line to enable the relative contribution of the different radionuclides…

…present in the gas to be determined. The Hitachi criticality monitor is connected in series for simultaneous calibration.

^{85m}Kr, ^{87}Kr, ^{88}Kr and ^{135}Xe from ^{235}U

A gamma-ray energy spectrum of the extracted FP gases is shown in fig. 4. Photopeaks of ^{85m}Kr, ^{87}Kr, ^{88}Kr, ^{88}Rb, ^{133}Xe and ^{135}Xe were identified. All other peaks present in the spectrum were attributed to NORM present in the fabric of the building. No evidence was found for other FPs or APs, which indicates that only noble gases were released from the target assembly during the gas extraction process. The only non-noble gas present was ^{88}Rb. As a short-lived daughter of ^{88}Kr, ^{88}Rb rapidly re-establishes transient equilibrium with ^{88}Kr following initial extraction.

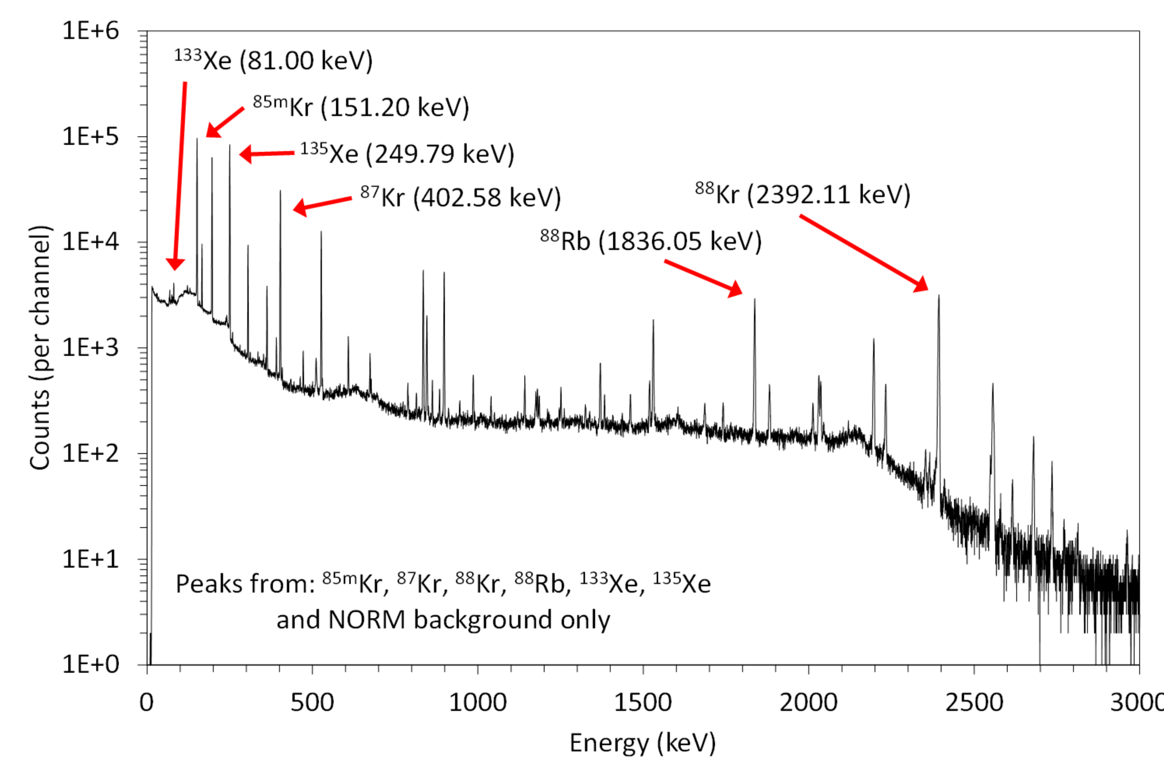

Fig. 4 – Gamma-ray energy spectrum of extracted FP gases.

A total gas activity of 12.3 kBq was extracted (referenced to start of measurement; 4 h after the end of irradiation). The relative contribution of ^{85m}Kr, ^{87}Kr, ^{88}Kr, ^{88}Rb and ^{135}Xe was 2.0, 1.6, 3.3, 3.4 and 2.0 kBq, respectively.

^{41}Ar from natural argon

A gamma-ray energy spectrum of the AP gas ^{41}Ar extracted from the natural argon target is shown in fig. 5. A total activity of 15.7 kBq was produced (referenced to end of irradiation).

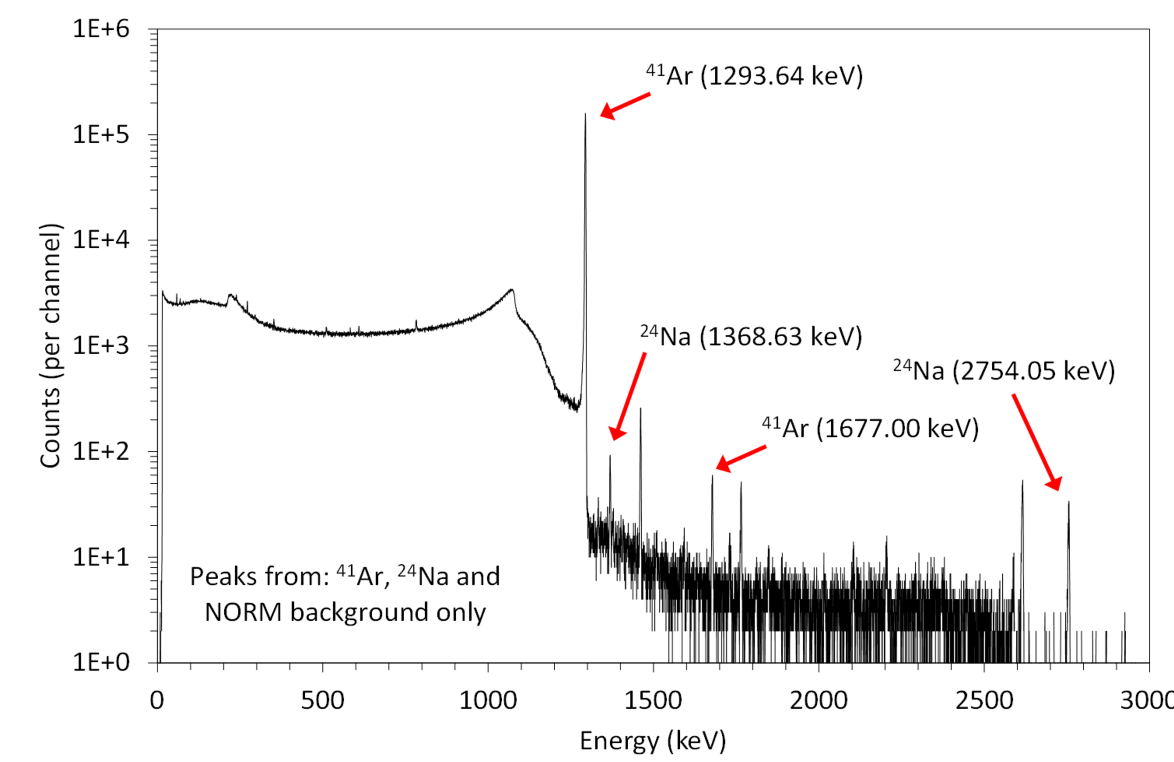

Fig. 5 – Gamma-ray energy spectrum of ^{41}Ar. Trace levels of ^{24}Na attributed to ^{23}Na contamination on the target case.

Summary and future work

Methods for the production, extraction and measurement of the short-lived FP/AP gases ^{41}Ar, ^{85m}Kr, ^{87}Kr, ^{88}Kr and ^{135}Xe have been developed. A trial calibration has been completed of a prototype criticality monitor developed by Hitachi and Hitachi-GE for the Fukushima NPP recovery programme.

Work is on-going to extended the methods developed through this project to produce CTBTO-relevant gas standards. This includes delayed 'milking' of the ^{235}U target to extract ^{133}Xe and irradiation of natural xenon to produce a range of xenon radioisotopes.

Acknowledgements

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