Design and Testing of a Combined Neutron and Gamma Assay System for Measuring Fissile Material in Fuel and Waste Items -15340

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ABSTRACT

A combined neutron and gamma ray assay system for the measurement of fissile material in both fuel and waste items has been designed, modelled, developed and tested. The purpose of the system is to measure and characterize a wide range of fissile materials including uranium, plutonium, americium and mixed oxide fuel arising from a variety of processes and facilities across the United Kingdom. Dounreay Site Restoration Limited at Dounreay currently stores these items form the United Kingdom Nuclear Decommissioning Authority's un-irradiated fuel inventory. This inventory comprises historic materials from ex-United Kingdom Atomic Energy Authority nuclear fuel projects and experiments together with fast reactor fuel materials and some commercial fuel items as delivered to Dounreay for recovery. The combined assay system has been designed and constructed to support the operation of a fuel processing facility, which will be operated at Dounreay to treat these historic legacy fuel materials, making them suitable for long term storage. Appropriate characterization of the fissile material is required for safety, criticality control and nuclear materials safeguards purposes prior to final disposition. The system consists of both gamma ray and neutron measurement components. The results of a neutron measurement are combined with the gamma ray isotopic ratio measurement results to generate the assay result for the fissile material being characterized. The gamma ray component of the system employs a safeguards quality high purity Germanium detector with electro-mechanical cooling and the measurements of fissile material containers are made in a shielded chamber with a tin and copper graded lining. Containers of fissile material are rotated during the gamma ray measurement and a variable automatic steel collimator aperture mechanism is employed to adjust the measured count rate to control detector dead time. Gamma ray spectra are analysed using the plutonium and uranium isotopic ratio code PC/FRAM. The gamma ray system can also be configured to perform a direct assay of small quantities of uranium bearing material. For this assay measurement a series of cadmium filters can be positioned in front of the detector to reduce the contribution from 59 keV gamma rays from Am-241. In addition to gamma ray isotopic measurements, both active and passive neutron assay is performed by the system. The neutron-measuring component consists of a thick walled cylindrical polyethylene chamber, which provides neutron moderation for 30 one-inch diameter He-3 detector tubes located in re-entrant channels in the polyethylene moderator. Two removable polyethylene-inserts incorporating lower plug assemblies are provided, one with and one without a cadmium lining. Also two top plug units are provided, again one with cadmium and one without a cadmium liner. The Neutron Subsystem can be operated both as a passive and as an active neutron coincidence counter. In both fast and thermal active neutron mode, the neutron component employs two americium-beryllium (Am-Be) alpha-n neutron sources, one positioned in the top and the other in the bottom plug unit. The neutron sources are removed when the counter is operated in passive mode. Passive neutron detection efficiency and active neutron performance has been modelled using the MCNP Monte Carlo computer simulation code as part of the design process. In the absence of fissile samples,

operation of the gamma ray component of the system has been tested initially using calibrated Eu-152 and Ba-133 gamma ray sources and the use of archived spectra from fissile material measurements. Initial testing of the neutron system has employed a calibrated Cf-252 spontaneous fission neutron source. Neutron testing has been confirmed using further MCNP simulations. The results of test measurements with the system are presented and compared with the results of MCNP measurement simulations.

INTRODUCTION

The integrated Fissile Assay System (FAS) is capable of assaying a range of fissile materials such as uranium of different enrichments, plutonium, americium and mixed oxide (MOX) fuel within various matrices. The results obtained will be used as the definitive nuclear material declaration value of each individual package for safety, criticality control and nuclear materials safeguards purposes. Some of the inventory was previously measured [1] as it was transferred to Dounreay Site Restoration Limited (DSRL) at the Dounreay site. The United Kingdom Atomic Energy Authority (UKAEA) previously operated the site.

Two instrument sub-systems make up the integrated instrument. The first is a Neutron Assay active and passive neutron coincidence counter (ANCC/PNCC) Subsystem for the measurement of fissile material present in product packages and waste containers. The second is a Gamma Assay Subsystem to provide a complete isotopic inventory of all actinides (primarily plutonium, uranium and americium) present in each package or container. The Gamma Assay Subsystem is also able to carry out an assay of waste containers of low activity uranium material. Finally, a Workstation, which is connected to both the Neutron and Gamma Assay Subsystems, houses the main computer and provides a User Interface (UI) for operation of both subsystems and the combining of data from neutron and gamma measurements to provide a complete inventory of all actinides present in the packages.

The Gamma Assay Subsystem is used for plutonium and uranium isotopic analysis of gram to kilogram quantities of material and the assay of waste containers of low activity uranium material. It operates in both Isotopic Analysis Mode and Assay Mode. In Isotopic Analysis Mode the system measures a package to determine the complete isotopic inventory of all actinides (primarily plutonium, uranium and americium). In Assay Mode the system is used to conduct an assay on low activity Uranium waste that will not be measured by the Neutron Assay System.

The Neutron Assay (ANCC/PNCC) Subsystem measures the mass of material present in fuel cans and waste containers by conducting passive and/or active measurements on all items, apart from low activity uranium waste, to determine the Special Nuclear Material (SNM) present in the item. It provides the capability to detect and measure:

    a.   Sub-gram quantities of plutonium in plutonium waste packages
    b.   Kilogram quantities of plutonium in Pu/MOX fuel items
    c.   Kilogram quantities of Enriched Uranium (EU) in fuel items, and
    d.   Gram quantities of EU in waste items.

The Neutron Assay Subsystem has three modes of operation:

    1.   **Passive Mode** to facilitate quantitative measurement of the plutonium-240 effective (^{240}Pu$_{eff}$) content of Pu/MOX fuel and waste items when operating in PNCC assay mode,
    2.   **Fast/Active (neutron interrogation) Mode**, and

3. **Thermal/Active (neutron interrogation) Mode** to facilitate quantitative measurement of EU fuel and waste items when operating in ANCC assay mode.

The information from a neutron measurement is combined with the isotopic information from the Gamma Assay System to provide a complete isotopic based inventory of all actinides present in each individual package or container. The sample measurement time for each instrument is typically between 10 and 20 minutes. The analysis software terminates a measurement at a pre-set time configured by the user, however, should the accuracy not be reached within this period, the count time is extended for an additional pre-set time. The theoretical basis of the neutron counting process is described elsewhere [2,3] and is based on neutron multiplicity theory developed by W. Hage.

The system Workstation consists of an enclosure, which houses the system main computer and the Model 1003 Time Correlation Analyser (TCA). The TCA is used to control the Neutron Subsystem and acquire neutron measurement data. The system Workstation is fitted with a worktop on which an LCD monitor and keyboard/mouse are located. A gland plate at the rear of the cabinet provides connection points for power to the Workstation, power and signal to the Neutron Assay Subsystem, signal from the Gamma Assay System and an High Voltage data take-off point for the Neutron Assay System for EURATOM safeguards purposes.

The main user interface for the system is the ANTECH MCC (Master Control Computer) software package, which is accessed by the user from the Workstation and is used to control the measurement of samples made by each subsystem instrument. It provides an interface for the user to select the type of measurement that is to be performed. Four different types of measurement, including active or passive neutron measurement, gamma isotopic or gamma assay measurement may be selected. These measurements are accomplished using respectively, ANTECH *MasterPassiveActive* – passive and active neutron counting software application, and ANTECH *IsoScan* gamma isotopics software employing the PC/FRAM code [4] and assay software. The MCC software controls and reports the progress of each measurement. It also combines the data from neutron and gamma ray measurements and presents the measurement results.

Both gamma and neutron detectors' signals provided to EURATOM will be acquired by unattended measurement software RADAR and independently analysed by the iRAP raw measurement data analysis program [5]. Other relevant references are also cited [6, 7, 8, 9].

DESIGN

Several parameters and constraints have been considered in order to arrive at designs for both the gamma and neutron subsystems to optimise the performance of the instrument for a range of fissile materials. In some cases conflicting physics requirements have had to be addressed. Allied to this conflict is the further requirement to be able easily to configure the system to carry out different measurement types for the different fuel and waste package sizes.

The main physics design considerations for the Gamma Subsystem design were:

a) Selection of High Resolution Gamma Spectrometry (HRGS) High Purity Germanium (HPGe) detector type and counting electronics.
b) Confirmation of the thickness of the graded lead shield for the measurement chamber to reduce the effect of external background radiation and supress re-emitted X-rays.
c) Limit the detector field of view of a sample with a very high gamma ray emission rate, in order to reduce the detector 'dead-time' and thereby minimise the count time required to reach the required accuracy.

d) Establish the thicknesses for a range of cadmium filters to be placed between the sample and HRGS detector to reduce, in particular, the flux of the 59 keV gamma rays arising from Am-241 to acceptable levels for both isotopic analysis and uranium assay. The Am-241 signal is the principle contributor to detector 'dead-time' for the measurement of plutonium.
e) Provide the capability to measure low activity U-235 and U-238 samples of up to 10g that are not measured by the neutron system.

A comprehensive physics assessment of the materials to be measured, the key parameters associated with these materials, and the measurements to be carried out for both passive and active neutron mode measurements in order to meet the performance requirements for the Neutron Subsystem was conducted. MCNP Monte Carlo modelling was used to design the Neutron Subsystem employing the MCNP 4C code running under Linux using the DXTRAN formalism and direction biasing. The design evolved through a step-by-step process by considering:

a) The dimensions of the counting chamber well.
b) The design of the removable cadmium and non-cadmium lined sleeves and end plugs with, or without, interrogation sources to accommodate different container sizes for waste and fuel.
c) The number and position of the He-3 detectors to achieve the required efficiency of 30% (required to meet the measurement time).
d) The detrimental effects on the He-3 detectors of the high gamma dose, in particular from Am-241 and proposed mitigation measures.
e) The external shielding required to reduce the effect of the external background.
f) The use of a pair of Am/Be interrogation sources for active measurements rather than Am/Li, due to the unavailability of the latter, and determination of the sources strength, position above and below the sample being interrogated and the use of moderation between the source and sample to tune the response of the system, and
g) The MDA for U-235 and PuO_2, and the detection efficiency for MOX with enriched Uranium, and Pu and U metals.

Of particular importance to the He-3 detectors is the effect of the high gamma dose-rate. It is important to ensue that the detectors are not exposed to a gamma radiation field greater than 1 Rad/h (equivalent to 10mSv.h^{-1} for gamma rays). The major contributor to the dose-rate is the Am-241 content of the PuO_2. The assessment confirmed that the expected photon dose-rate is well below this limit, however, additional measures were taken to reduce further the dose-rate.

CONFIGURATION OF THE FISSILE ASSAY SYSTEM

The final Gamma Subsystem design consists of:

a) A 25% efficiency ORTEC SGD-GEM-5050P4 HPGe detector and ORTEC DSpec Junior. (The detector amplifier supports 4 signal outputs; two primary and two secondary. One primary output feeds the ORTEC DSpec Junior 2.0. Two outputs, a primary and a secondary, are supplied for EURATOM Safeguards equipment for safeguards purposes. The remaining secondary output is used to control the variable aperture collimator for dead time reduction).
b) A 2.5-inch thick lead shielded sample measurement chamber with a copper and tin graded inner liner.
c) A turntable, onto which the sample to be measured is placed. This is to ensure any sample density variations have a minimal effect on the result.
d) An automatically adjustable steel collimator with a variable aperture consisting of two thick walled steel 'curtains' driven by a servomotor and controlled by the Gamma Subsystem software. The software and shutter controller monitor the detector count rate using the

detector amplifier secondary output and automatically move the 'curtains' to the most optimum position for the measurement to reduce the HPGe detector 'dead time'.

e) A set of three cadmium filters; 2, 5 and 8mm thickness, for manual insertion between the sample and the detector to further reduce the intensity of the of the 59 keV gamma ray line from Am-241, where appropriate.

The Gamma Subsystem can also conduct an assay on low activity uranium waste that will not be put through the neutron assay system. For this mode of operation the adjustable collimator is configured to travel to a set position and remain in this position for the duration of the assay. The Gamma ray Subsystem requires an energy and efficiency calibration for this mode of operation. The configuration of the Gamma ray Subsystem is shown in Figure 1.
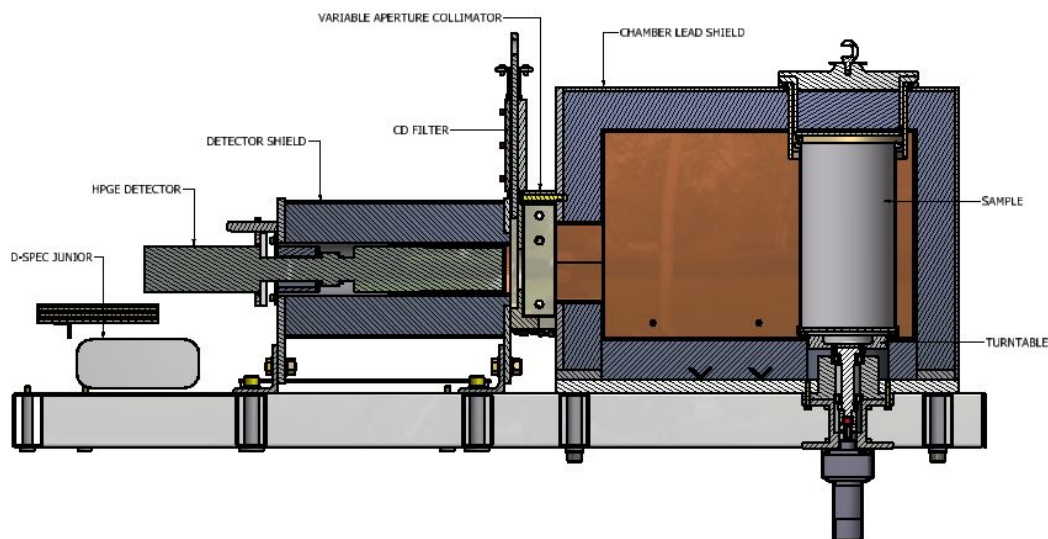

Fig. 1.   Cross-section view of the Gamma ray Measurement Subsystem.

The final Neutron Subsystem design consists of:

a) A chamber body manufactured from High Density Polyethylene (HDPE) moderator, with a central cylindrical measurement chamber well.
b) An inner sleeve (Insert), located within the chamber, which provides close coupling to the measurement sample to maximise the efficiency of the chamber. The insert can be removed through the top of the chamber by lifting with the aid of a hoist. A choice of two inserts is provided: one for 'Fast' neutron interrogation mode the other for 'Thermal' neutron interrogation mode. Both inserts are constructed from polyethylene: the 'Fast' mode insert is lined with cadmium clad in a protective steel lining and the 'Thermal' mode insert is similar in geometry but does not contain cadmium.
c) A lower end plug is built into the insert for ease of removal and safety, i.e. not requiring removal from underneath. The upper plug unit drops into the insert. Both the 'Fast' and 'Thermal' inserts and their corresponding top end plugs have the capability to position an Am/Be interrogation source for 'active' interrogation measurements.
d) 30 He-3 detectors arranged in a double ring and positioned to optimise the performance for both passive and active measurements.
e) A 3mm steel inner chamber liner to reduce the Am-241 contribution with the He-3 detectors lined on their inner surface with a magnesium coating to enhance their life (against a high gamma dose).

f) 70mm of high density polyethylene fitted around the outside of the chamber body with a 1mm cadmium liner positioned between the external shielding and the outside of the chamber body to ensure background thermal neutron flux is intercepted and absorbed prior to reaching the He-3 tubes.

g) A pair of Am/Be interrogation sources with an output of 2 x 10^5 n/s positioned above and below the sample.

h) Optimisation of the position of the Am/Be sources with respect to the sample centre point and the cadmium liner installed above and below the end plugs and positioning of a suitable thickness of polyethylene moderator between each source and the sample to reduce the minimum detectable activity (MDA) for the measurement of uranium samples.

All He-3 detectors are connected to a cylindrical junction box containing 6 Amptek A-111 high-count rate charge sensitive amplifiers. These are connected to a local enclosure which houses a De-randomising Mixer Buffer Counter (DMBC) to combine each amplifier input and provide a single output pulse train to the TCA. A secondary identical output is provided for connection to the EURATOM Safeguards equipment for independent data verification. Additional electronics distribute low and high voltage dc power to the junction box. The Neutron Measurement Subsystem is shown in Figure 2.
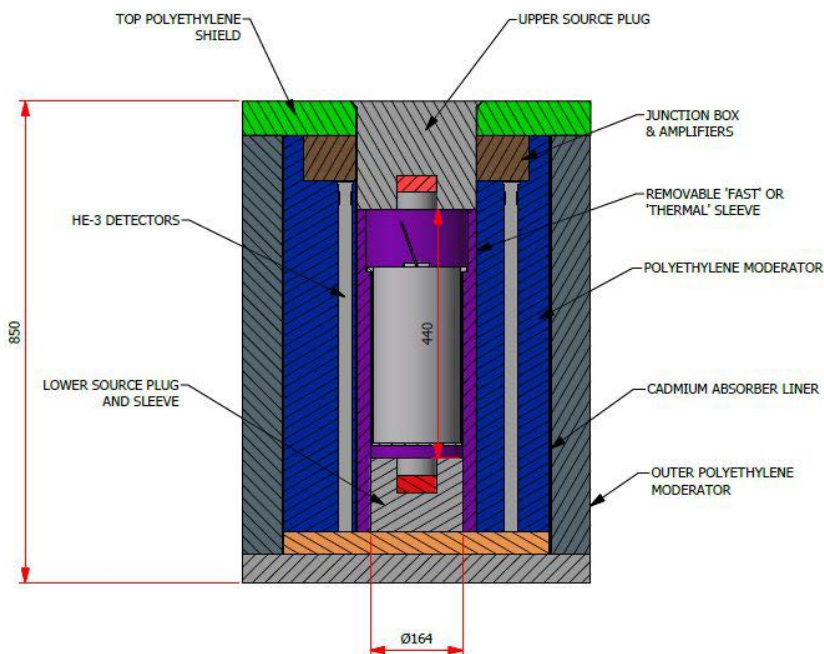

Fig. 2. Cross-section view of the Neutron Measurement Subsystem.

RESULTS OF PRELIMINARY TEST MEASUREMENTS

A number of design parameters have been confirmed as part of preliminary testing prior to testing with fissile material on the DSRL site. The first of these tests involved an independent corroboration of the outputs from both the Gamma ray and Neutron Subsystem counting electronics by connecting both ANTECH and EURATOM counting electronics and software to a duplicate set of data outputs, (provided for safeguards verification measurements).

The Gamma ray Subsystem test employed the 25% efficiency ORTEC SGD-GEM-5050P4 HPGe detector and amplifier, which provide 4 signal outputs: two primary and two secondary.

One primary output feeds the ORTEC DSpec Junior 2.0 supplied with the system. Two further outputs, a primary and a secondary, are supplied for EURATOM Safeguards purposes. The primary output supplied for ANTECH data analysis purposes is analysed with PC/FRAM isotopic ratio analysis code. The primary and secondary data outputs for EURATOM were processed by a mini-MCA.

TABLE I. Comparison of HRGS detector parameters measured simultaneously by ANTECH and EURATOM Safeguards employing duplicate outputs. Measured parameters are highlighted in bold in the table. [Sources used include Ba-133, 769 kBq (748 kBq on 3 September 2014) and Eu-152, 829 kBq (811 kBq on 3 September 2014].

Parameter and Results	EURATOM Primary	EURATOM Secondary	ANTECH
Polarity of signal	Positive	Positive	Positive
Shaping time	High	High	High
Coarse Gain	200	200	200
Fine Gain	1.2388	1.2388	1.2388
PZC	1782	1782	1782
Counting time, S	15	15	15
Measurement time LT, S	**622**	**637**	**631**
Dead-time, %	**1.27**	**1.24**	**1.25**
Integral of counts	**2.62E+05**	**2.67E+05**	**2.63E+05**
Rotation of can	same	same	same
Cd filter	same	same	same
Aperture	same	same	same
FWHM, eV (for 122 keV line)	**758**	**810**	**797**

Verification of the performance of the Neutron Subsystem included measurement of neutron detection efficiency with and without cadmium moderator and the dead-time of the system, using calibrated Cf-252 sources and independent corroboration of the outputs from the Neutron Subsystem counting electronics by parallel connection of both ANTECH and EURATOM counting electronics and software. These results are presented below.

The Neutron Subsystem measurements were demonstrated for Passive Mode operation with a Cf-252 source positioned centrally and at the mid-point of the He-3 detectors active length for both cadmium and non-cadmium liners. The detection efficiency exceeds the design requirement of 30% for both conditions. ANTECH neutron measurements were made with the

ANTECH Model 1003 Time Correlation Analyser and *Master PassiveActive* software. The data is presented in TABLES II and III. EURATOM measurements (TABLE III) were made with Canberra JS-15 counting electronics and software.

TABLE II. Passive Mode Efficiency with and without Cadmium Liner (ANTECH measurement). [HV=1740V, PD=4µS, GW=64µS, t=1200 seconds, cycle length=30 seconds. ^{252}Cf source 21543B: 2549 n/s on 3 September 2014].

^{252}Cf Source No.	Reference Activity (n/s)	Reference Date	Half Life (days)	Measured date	Decay Corrected Source Neutron Output (n/s)	Cd Liner	Measured Neutron Output (n/s)	Point source with no matrix Efficiency
21543B	2930	21-Feb-14	966	09-Jul-14	2679	Present	821.3	30.66%
21543B	2930	21-Feb-14	966	10-Jul-14	2679	Absent	898.1	33.53%

TABLE III. Passive Mode Efficiency with Cadmium Liner (EURATOM measurement). [HV=1740V, PD=4µS, GW=64µS, t=4600 seconds, cycle length=100 seconds. ^{252}Cf source 21543B: 2549 n/s on 3 September 2014].

^{252}Cf Source No.	Reference Activity (n/s)	Reference Date	Half Life (days)	Measured date	Decay Corrected Source Neutron Output (n/s)	Cd Liner	Measured Neutron Output (n/s)	Point source with no matrix Efficiency
21543B	2930	21-Feb-14	966	03-Sep-14	2549	Present	813.74	31.74< 31.91 <32.08%

The Totals and Real count rates for both the ANTECH and EURATOM neutron system outputs agree well within the accuracy with which the count periods can be synchronised. This data is presented in TABLE IV.

TABLE IV. Comparison of EURATOM and ANTECH measured neutron count rates. [HV=1740V, PD=4µS, GW=64µS, t=2110 seconds, cycle length=10 seconds. ^{252}Cf source 21543B: 2549 n/s on 3 September 2014].

Measurable Value	EURATOM	ANTECH
Totals, cps	813.2±12.3	812.2±1.2
Reals, cps	243.1±8.9	244.2±0.6

The Die-away time calculated as part of the MCNP Monte Carlo modelling of the system was 65.11µs. The die-away time measured using the Antech counting electronics and employing the Hage Point Kernel Method [2] was 67.6µs. The die-away time measured independently by EURATOM Safeguards using two gate widths of 64µs and 128µs was 65µs. This data is presented in TABLE V.

TABLE V. Die-away Time. [HV=1740V, PD=4µS, cycle length=100 seconds. ^{252}Cf source 21543B: 2549 n/s on 3 September 2014].

Value	GW$_1$=64µS t=4600 sec.	GW$_2$=128µS t=2200 sec.
Reals, (cps)	242.78±3.09	333.48±3.75
$\tau = \dfrac{-64}{ln\left(\frac{Reals_{128}}{Reals_{64}}-1\right)}$, µS	60.87 < **65.00** < 69.13	

In TABLE VI below, a measurement is presented of the Reals to Totals ratio for the Neutron Subsystem.

TABLE VI. Estimation of ρ_0. [HV=1740V, PD=4µS, GW=64µS, t=4600 seconds, cycle length=100 seconds. ^{252}Cf source 21543B: 2549 n/s on 3rd September 2014]. Note that k$_{cor}$ is the coefficient for correction of differences of factorials moments of ^{252}Cf and ^{240}Pu and their efficiency. $k_{cor} = \dfrac{3.757 * 3.825}{2.156 * 11.962} * 1.02$

Value	TOTALS	REALS
Counts per second	813.47±4.30	242.78±3.09
$\rho_0 = \dfrac{Reals}{Totals} * k_{cor}$	0.167 < **0.170** < 0.172	

CONCLUSIONS

This paper describes the design and presents preliminary performance results for a Fissile Assay System developed for measuring and characterising the UK inventory of un-irradiated exotic nuclear fuel stored by Dounreay Site Restoration Limited. The design considerations required to optimise the systems for the wide range of materials and matrices have been discussed. Preliminary testing of the system has been conducted at ANTECH to confirm agreement between MCNP modelling data and measured data for neutron detection efficiency and die away times for passive mode operation.

Tests conducted show good correlation of the output data from both the neutron and gamma systems counting electronics and equivalent electronics and counting software connected to a duplicate set of additional data outputs provided for independent safeguards measurements by EURATOM.

Calibration and final verification of the system performance will be undertaken using a sub-set of well characterised fuel samples (standards) at the Dounreay site so that the FAS can be used to characterise the expected range of fuel types which constitute the inventory.

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