# DEVELOPMENT, TESTING AND VALIDATION OF A WASTE ASSAY SYSTEM FOR THE MEASUREMENT AND CHARACTERISATION OF ACTIVE SPENT FUEL ELEMENT DEBRIS FROM UK MAGNOX REACTORS

John A. Mason*, Kevin J. Burke*, Marc R. Looman*, Antony C. N. Towner* and Martin E. Phillips**
*ANTECH, A. N. Technology Ltd., Unit 6, Thames Park, Wallingford, Oxfordshire, OX10 9TA, UK
** Nympsfield Nuclear Ltd, Chapel House, The Cross, Nympsfield, Stonehouse GL10 3TU, UK (formerly with Waste Management Group, British Nuclear Group)

## ABSTRACT

This paper describes the development, testing and validation of a waste measurement instrument for characterising active remote handled radioactive waste arising from the operation of Magnox reactors in the United Kingdom. Following operation in UK Magnox gas cooled reactors and a subsequent period of cooling, parts of the magnesium-aluminium alloy cladding were removed from spent fuel and the uranium fuel rods with the remaining cladding were removed to Sellafield for treatment. The resultant Magnox based spent fuel element debris (FED), which constitutes active intermediate level waste (ILW) has been stored in concrete vaults at the reactor sites. As part of the decommissioning of the FED vaults the FED must be removed, measured and characterised and placed in intermediate storage containers. The present system was developed for use at the Trawsfynydd nuclear power station (NPS), which is in the decommissionning phase, but the approach is potentially applicable to FED characterisation at all of the Magnox reactors. The measurement system consists of a heavily shielded and collimated high purity Germanium (HPGe) detector with electro-mechanical cooling and a high count-rate preamplifier and digital multichannel pulse height analyser. The HPGe based detector system is controlled by a software code, which stores the measurement result and allows a comprehensive analysis of the measured FED data. Fuel element debris is removed from the vault and placed on a tray to a uniform depth of typically 10 cm for measurement. The tray is positioned approximately 1.2 meters above the detector which views the FED through a tungsten collimator with an inverted pyramid shape. At other Magnox sites the positions may be reversed with the shielded and collimated HPGe detector located above the tray on which the FED is measured. A comprehensive Monte Carlo modelling and analysis of the measurement process has been performed in order to optimise the measurement geometry and eliminate interferences from radioactive sources and FED in the immediate vicinity of the measurement position. The detector system has been calibrated and high activity radioactive sources of Cs-137, Co-60 and Na-22 have been used to validate the measurement process. The data acquisition and analysis software code has been tested and validated in keeping with the software quality assurance requirements of both ISO:9001-2008 – TICK-IT in the UK and NQA-1. The measurement and analysis system has been comprehensively tested with high activity sources, is flexible and may be applicable to a wide range of remote handled radioactive waste measurement applications.   It is due to be installed at Trawsfynydd NPS later this year.

## INTRODUCTION

Magnox based spent fuel element debris (FED), which constitutes active intermediate level waste (ILW) has been stored in concrete vaults at the various Magnox reactor sites. FED consists of the magnesium-aluminium alloy based cladding and splitters

which maintained the position of the fuel elements in the core channels. The cladding and splitters were stripped in the fuel cooling ponds to enable better packing of the fuel being transported for reprocessing. The process often damaged the fuel elements, causing the end fittings containing high-cobalt nimonic springs to become incorporated in the fuel element debris. The FED picked up fuel contamination from the cooling ponds, and sometimes fuel fragments (or larger pieces of fuel) adhered to the end fittings. As part of the decommissioning of the Magnox reactors and FED vaults the FED must be retrieved from the vaults, measured and characterised to determine its radiological content and placed in approved containers (originally 3 $m^3$ boxes prior to grouting) for intermediate storage.

The challenge for the assay system is to be able to determine the content of Cs-137, and in turn to enable the fissile burden to be assessed using a radionuclide fingerprint, in the presence of higher and highly variable quantities of Co-60 from nimonic springs. Therefore, to improve the Cs-137 detection capability and hence the speed of waste throughput, the original low-resolution assay system was to be replaced. The dynamic range, in the sense of the amount of Cs-137 that can be quantified in the presence of higher levels of Co-60 is much better using a high-resolution assay system. In turn the use of HRGS minimises the amount of operator intervention required to locate and remove nimonic springs when too many are present on a tray to enable adequate quantification of Cs-137. A further requirement was for the software to be easy to operate and to maintain an inventory of material placed into each box of waste. The software also needs to inform the operator if intervention is needed to sort for fuel fragments or excessive levels of Co-60 bearing items.

The HRGS based waste assay system [1] utilises a tray counting geometry located in the retrieval plant Sort Cell. The detector and collimator are located beneath the assay tray in a nominally clean, uncontaminated area. Adequate system counting performance is achieved using a high efficiency detector with a suitably designed collimator and gamma filters and high count rate nucleonics.

The waste assay system includes a hyper-pure germanium detector with electro-mechanically cooled cryostat. The detector system is heavily shielded by lead and incorporates a tungsten collimator and stainless steel gamma-ray filters. The spectral data is analysed using a multi-channel analyser (MCA) and bespoke spectrum analysis software. Also included in the system are cabling, nucleonics, computer and printer, equipment cabinet, and uninterruptible power supply (UPS).

The measurement characteristics of the system have been modelled using MCNP Monte Carlo modelling [2] which was used to estimate measurement performance and eliminate sources of interference that might degrade the measurements. Measurement errors [3] have also been estimated through the modelling. The system has been extensively tested using intense gamma ray sources.

**MONTE CARLO MODELLING**

Extensive use was made of the MCNP Monte Carlo modelling procedure as an important component of the design process.The geometry of the assay cell was modelled including the details of the HPGe detector crystal, detector shielding and collimation and the waste distributed on an assay tray. The calculations, particularly including the detector response function, were confirmed against a benchmark measurement using radioactive sources.
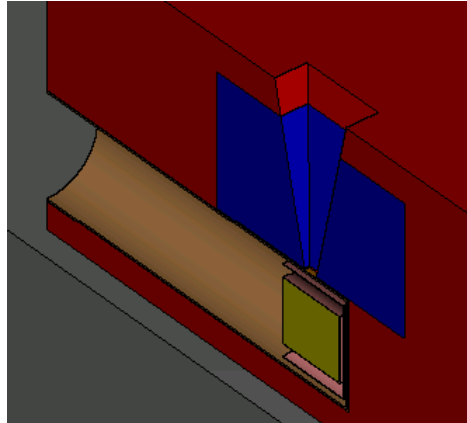
Figure 1. 3D geometry plot of the MCNP model showing vertical section through the HPGe detector crystal. The crystal is located at the center of the lead shielding (red coloured). The tungsten collimator (blue coloured) has an inverted pyramid shape, in order to view the entire tray with nuclear waste.

During the testing phase of the assay system comparisons were made between MCNP simulations and measured results. An example is displayed in Table 2, where experimental and simulation results for the measurement of a Co-60 point source with an activity of 25 GBq are compared.

In order to reduce the time required to perform MCNP calculations for multiple cases with complex geometry ANTECH have implemented a 20 processor computer cluster to achieve efficient parallel processing.

**MEASUREMENT HARDWARE**

ANTECH have developed an heavily shielded High Resolution Gamma Ray Spectroscopy system for the assay of active Intermediate Level Waste (ILW). It has application to a variety of remote handled (RH) waste measurement applications. As described in the present paper, the assay system is configured for and is applied to the measurement of waste trays containing retrieved Magnox FED waste. In this configuration it is designated as the Waste Tray Assay System (WTAS).

At the centre of the system is a 30% HPGe ORTEC PopTop detector with transistor reset pre-amplifier and high voltage filter. The PopTop style detector was chosen as it is suitable for connection to the Ortec X-Cooler, electro-mechanical cooling unit which allows the system to avoid the use of liquid nitrogen. A digital gamma ray spectroscopy system (Ortec DSPEC) is used for spectral data acquisition. It is housed outside the cell in the Operations Area and located within an environmentally controlled equipment rack.

The HPGe detector is located in a graded lead (Pb-Sn-Cu) shield and tungsten (W) collimator assembly, which is in turn housed in a stainless steel frame secured to a movable trolley. All exposed Pb is clad in stainless steel to avoid skin contact and to facilitate decontamination in the unlikely event that this becomes necessary. The Pb shielded and W collimated detector is located so that the side of the HPGe detector crystal views the waste from beneath the waste tray through a 2 mm thick stainless steel contamination protection plate, as illustrated in Figures 2 and 3 below.  The Pb shield and W collimator provide shielding for the detector not only from the waste on the tray but also from gamma radiation arising in the adjacent NIREX box, into which the waste

on the tray is transferred following the measurement.   Radiation from the NIREX box is scattered from the sorting cell roof.   The detector design demonstrates the importance and utility of MCNP modelling to ensure that all sources or radiation are considered in the shield design.

The trolley is positioned underneath the Sort Cell within the Assay Area in a fixed location and geometrically aligned beneath the waste tray in the Sort Cell. The trolley rolls on wheels. On one side the wheels move along a HEPCO Vee-rail which provides alignment. On the other side the wheels run along a flat steel plate. A feature on the rear end of the shield trolley allows a hook or grab to be attached such that the complete assembly can be moved into the man-access Operations area by removal of the shield plug. A mechanism is included in the Vee-rail to precisely position the trolley in the measurement position, based on mechanical stops.

The detector cooling system transfer hose and cables are designed to pass under the plug unit in a shallow trench so that they can be installed and removed easily without interfering with the operation or movement of the plug. The PopTop detector is connected to the X-Cooler via the cryostat hose connection. The X-Cooler is located outside the cell in the Operations Area with an appropriate vibration reduction mounting. The cryostat transfer hose is designed to pass through a short shallow trench and pass at an angle under the cell stepped plug unit. Similarly the HV Bias and signal cables are routed from the detector shielded trolley in the Assay Area beneath the Sort Cell and are designed to pass through a short shallow trench under the cell stepped plug unit to the instrument rack housed outside in the Operations Area.
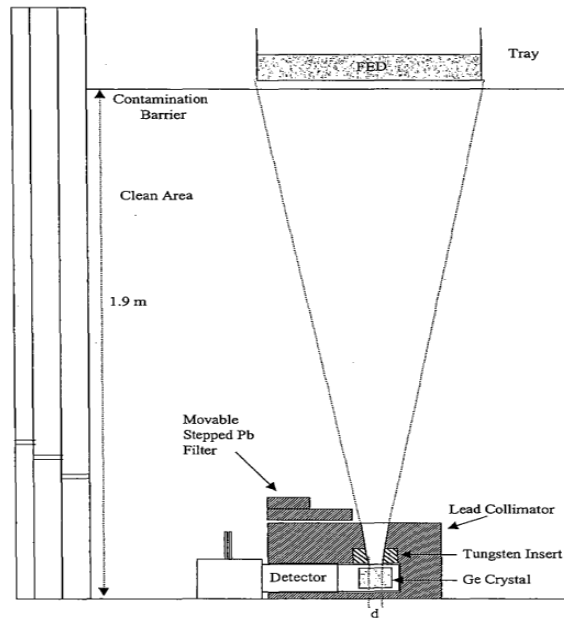


Figure 2. FED vault area showing shielded HPGe detector system below the FED-sorting tray. The biological shield with lower access door can be seen on the left.

The shielded detector/collimator trolley of the WTAS is shown in position underneath the Sort Cell in Figure 3. The containment plate above has been 'cut away' to show the imaginary 'field of view' of the waste tray above the collimated detector. The shield plug has been removed for clarity. The X-Cooler is shown outside of the cell with the transfer

hose connected at the rear of the HPGe detector. The trolley can be moved on the Vee-rail and removed from the cell for maintenance.
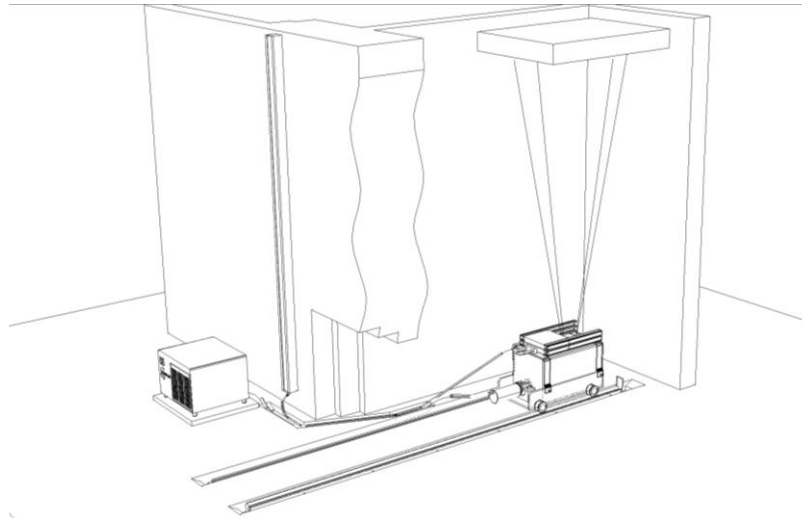


Figure 3. Artist concept view of the ANTECH Waste Tray Assay System (WTAS) beneath the Sort Cell showing the sorting tray and the HPGe electro-mechanical cooler external to the measurement cell.

Figure 4 is a photograph of the WTAS detector head and shielding with the stainless steel filters visible on the top of the unit.



Figure 4. Waste Tray Assay System (WTAS). The stainless steel filters and the filter motion control system can be seen on the top.

**ANALYSIS SOFTWARE**

The main data analysis and data acquisition component for the FED Waste Tray Assay System is the ORTEC GammaVision™ MCA software for multichannel analyser (MCA) control and data acquisition. The system also employs ANTECH bespoke software, based on appropriate ORTEC software tools, for spectrum analysis and nuclide activity waste assay calculations. The software is also capable of performing system calibration and maintenance functions. The software is designed to perform the following tasks:

1. Interface with the operator to initiate and control measurement sequences, and communicate with the plant control PLC, digital spectrometer and stainless steel gamma filter positioning system,
2. Measure the activity of gamma ray emitting radio-nuclides of interest in waste in a tray (Cs-137 and Co-60),
3. Measure or accept data entry for nuclides of interest in FED waste (i.e. the relationship between the measurement of fuel contamination, identification of fuel pieces, nimonic springs and the fingerprint data including the FED weight option),
4. Read the weight of the tray as an analogue input
5. Calculate, store, and display an inventory for a box (containing the contents of a number of measured trays) based on individual measurements and data entry,
6. Perform check measurements to confirm the correct functioning of the system,
7. Enable an Operator or Supervisor, under Supervisor password control, to access and edit configuration parameters, alarms settings, threshold settings, fingerprints, algorithm parameters etc,
8. Data backup,
9. Perform diagnostic and maintenance functions,
10. Generate of hard copy reports of Box inventory, fingerprint data, alarms and threshold settings.

The typical phases and features of the operation of the measurement process that are built into the software are listed below:

**Initial Count**

- Check the dead time against the lower and upper threshold (using pre-set thresholds)
- If Cs-137 is above the MDA Threshold (Bq), terminate the assay, or
- Predict full count time to reach Cs-137 MDA: if higher than the threshold display 'Warning'. In this case prompt the operator to remove nimonic springs, retry, continue or halt the measurement
- If the dead time is above the upper pre-set threshold, stop count, produce warning alarm and give options including override upon entry of supervisor password. Select next filter in sequence. Restart measurement
- Check the dead time against the upper threshold
- Select other steel filters as required
- When a filter is in place, provide a visual indication

**Main Count**

- If, at the end of the initial count period, the MDA warning message is not triggered, the main count will start and a) continue until the MDA is met (if this option is enabled) or b) acquire for the full count period.
- Display the tray results: Activity of principle radionuclides (Bq) and fissile content (g). Associated uncertainties shall be given. MDAs will be reported if the principle radionuclides are not positively identified.
- Tray accepted by operator or supervisor – end of measurement sequence.

**Print/Display Options**

- Display the gamma ray spectrum during the acquisition of counts - the vertical scale of the spectra will change automatically as the counts in the spectrum increase. The vertical scale is adjustable manually and there is a Log/linear scale option
- Start date and time of count acquisition
- Time to go to the end of the main count (Real time)
- Count to an MDA option
- Abort the measurement

**Alarms**

- Screen indication of the estimated 'time to reach set MDA'
- At the end of the measurement, compare dead time against high dead time threshold. Alarm if above and list options for operator.
- At the end of the measurement, compare Cs-137 MDA against Cs-137 MDA threshold. Alarm if above and list options for operator.
- At the end of the measurement, compare Cs-137 activity against fissile and high fissile alarm thresholds. Alarm if above and list options for operator.
- Compare box Co-60, Cs-137 and fissile content (before tray contents added) with pre-set box limits for Co-60, Cs-137 and fissile content. Alarm if above, and list options available to operator.

## WTAS PERFORMANCE AND TESTING

The WTAS is designed to measure average and maximum volumes of waste per tray of 25 and 50 litres respectively. The nominal design average (over the whole FED vault) of Co-60 activity in 25 litres of waste is $6.9 \times 10^9$ Bq while the maximum Co-60 activity in 50 litres of waste (excluding abnormal items) is $5.6 \times 10^{10}$ Bq. The system should measure at this level without deploying the stainless steel filters. In FED waste the Co-60 activity is expected to arise from loose nimonic springs evenly distributed throughout the waste. The nimonic springs are attached to spiders which are part of the top end fitting of Magnox fuel elements. They have a relatively high cobalt content which, with neutron activation, results in enhanced Co-60 activity. The FED on the tray (average density 0.35 g/cc) will have a typical depth of 10 cm.
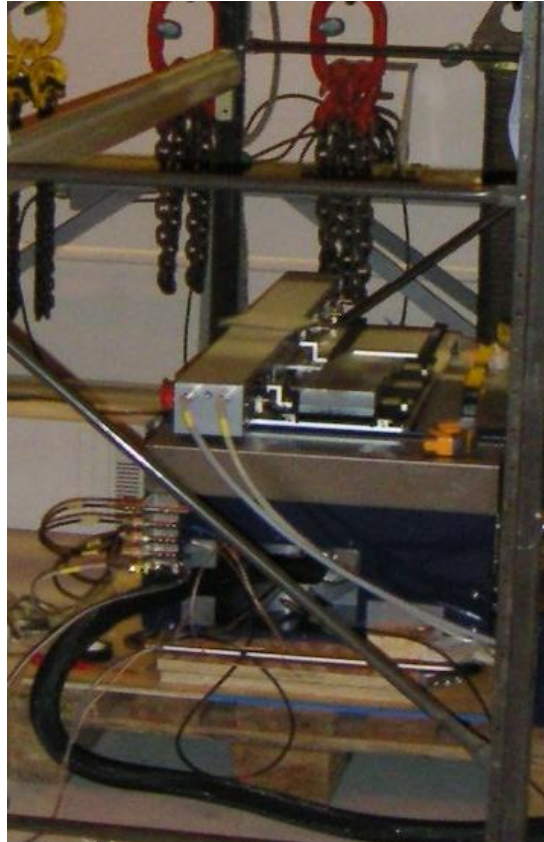
Figure 5. Waste Tray Assay System (WTAS) installed in an irradiation position during testing at the National Physical Laboratory in Teddington.

A further design feature is that the assay system must be capable of measuring 500 nimonic springs (4.2 x $10^{11}$ Bq of Co-60) or a 500 mm length of fuel element (maximum Cs-137 activity of 4.2 TBq) on the assay tray whilst staying within the count rate limit of the nucleonics. A thick gamma filter may be used to avoid the detector from saturating. The limit of detection (LOD) of the WTAS is also designed to be such that small pieces of fuel located in a waste tray can be detected in 50 litres of waste (0.35 g/cc) containing 5.6 x $10^{10}$ Bq of Co-60 activity. Assay times are expected to be less than or equal to 300 seconds.

Tests have been performed with radioactive sources at the Royal Military College of Science (RMCS) and at the National Physical Laboratory (NPL) to demonstrate system performance and to validate the operation of both the hardware and software. Figure 5 shows the WTAS with electrical cables and cryogenic hose connected in an irradiation position at the NPL.

The design of the detector geometry and collimation was tested by measurements performed at the NPL where sources were placed on a simulated waste tray located at a height of 155 cm. The measurements were performed to confirm the uniformity of the detector response to waste distributed on the tray. The measurement data is displayed in Table 1. The resulting variation in count rate across the waste tray is within the the design specification for the WTAS. The data is displayed in a contour plot in Figure 6.

|   | A | B | C | D | E | F |
|---|---|---|---|---|---|---|
| **1** | 5.6 | 6.0 | 6.2 | *6.2* | *6.0* | 5.5 |
| **2** | 6.6 | 6.8 | 7.0 | *7.0* | 6.9 | *6.7* |
| **3** | 7.0 | 7.8 | 7.8 | *7.9* | 7.7 | 7.2 |
| **4** | 7.3 | 7.8 | 8.0 | 7.9 | *7.5* | *6.8* |
| **5** | 7.9 | 8.3 | 8.0 | 7.9 | 8.2 | 7.9 |
| **6** | 8.0 | 8.2 | 7.9 | *7.4* | 8.2 | *7.7* |
| **7** | 7.5 | 7.8 | *7.4* | 6.5 | *8.0* | 7.6 |
| **8** | 7.2 | *7.3* | 6.6 | 5.3 | *7.6* | 7.4 |

Table 1. Measured count rates (s⁻¹) for a small point source placed at various X and Y grid positions of a simulated waste tray. The average measurement error is equal to 0.3 (s⁻¹) with a 95% confidence interval (2σ). The values in *italic* font are calculated with quadratic fits to the experimental data.

The dose rate range was tested by measuring the detection efficiency of the system with the detector exposed to strong radioactive sources that simulate measurements of FED. The measurements were conducted with strong Co-60, Cs-137 and Na-22 sources in an irradiation cell at RMCS. Data for the measurements with a Co-60 source are displayed in Table 2, for the case of no filter and with the three stainless steel filters with thicknesses of 24 mm, 42 mm and 56 mm. Also displayed in the table for comparison are the simulation data from a benchmarked MCNP Monte Carlo simulation. It can be seen that good agreement has been obtained between measured and simulated data.
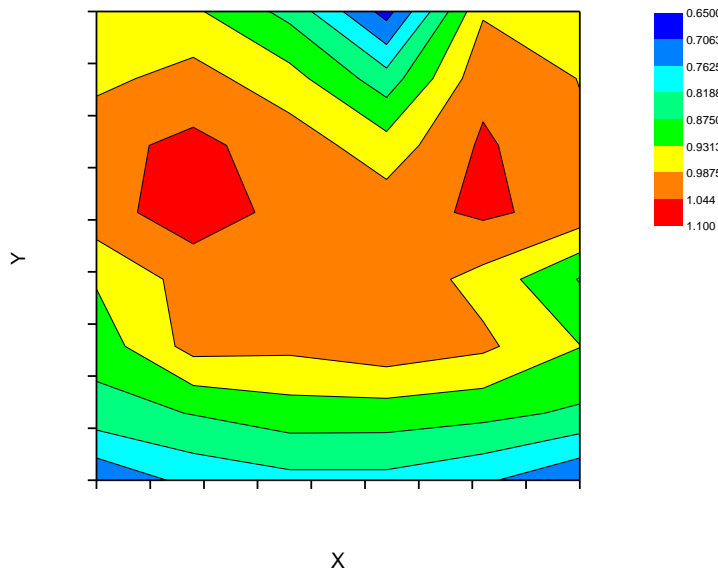


Figure 6. Contour plot of normalised experimental count rates. The values are normalised to the count rate of the central position on the tray, which is equal to 7.8 s⁻¹. The two small highest count rate regions are believed to be an artifact of the choice of boundary values and relatively large counting errors.

| Filter thickness (mm) | Efficiency | | | | | |
|---|---|---|---|---|---|---|
| | Integral Spectrum | | | Photopeak | | |
| | Exp | Sim | Sim/Exp | Exp | Sim | Sim/Exp |
| 0 | 3.6E-06 | 3.3E-06 | 0.93 | 2.6E-07 | 2.7E-07 | 1.06 |
| 24 | 1.4E-06 | 1.3E-06 | 0.91 | 9.9E-08 | 1.0E-07 | 1.04 |
| 42 | 6.9E-07 | 6.1E-07 | 0.88 | 4.8E-08 | 5.0E-08 | 1.02 |
| 56 | 4.1E-07 | 3.4E-07 | 0.85 | 2.8E-08 | 2.8E-08 | 1.01 |

Table 2. Experimental and simulation results for the measurement of a Co-60 point source with an activity of 25 GBq. The source is positioned centrally on the tray with a Pantatron Teleflex cable. The tray is located at a height of 155 cm. The simulation results were obtained with the MCNP4C2 code.


**CONCLUSIONS**

This paper describes the Waste Tray Assay System (WTAS) that has been developed for the measurement of Magnox FED waste. The WTAS has been tested with a range of radioactive sources and its operation has been simulated with benchmarked MCNP Monte Carlo calculations. The measurement software has been validated as has the operation of the system for a range of strong radioactive sources.  A system based on the design is due for installation and operation in 2012.

The system has application to the measurement of Magnox Fuel Element Debris (FED) waste at other Magnox reactor sites.   The major design objective of the WTAS that has been achieved is the ability of the assay system to determine the content of Cs-137, and in turn to enable the fissile burden to be assessed using a radionuclide fingerprint, in the presence of higher and highly variable quantities of Co-60, typically from nimonic springs.

The approach can be used in other Magnox FED waste configurations where the detector is located above the FED waste sorting tray and where the collimation is fixed below the detector and at a distance above the tray.  In this case, which has also been investigated, there are different shielding problems and mechanical support issues.   The extensive use of MCNP Monte Carlo modelling to simulate the geometry of the sorting cell and the distribution of radioactive sources has helped to ensure that all of the detector shielding requirements are addressed and suitable Cs-137 and Co-60 discrimination can be achieved.

The WTAS in its present form or in other configurations has relevance to the measurement of other active ILW and highly active RH waste. Examples include high activity RH LLW and RH TRU (Transuranic) waste as defined in the United States arising from both commercial nuclear and Department of Energy (DOE) operations. The analysis is able to analyse a range of radionuclides beyong those expected in the Magnox FED cases.

**REFERENCES**

1. Curtis, J. N. (2003), Trawsfynydd North FED: Feasibility of An Assay System Utilising High Resolution Gamma Spectrometry, BNFL ENVT. SERVS/ET/REP/TRA/0733/03/Issue 1.

2. Briesmeister, J. F. (2000). MCNP - A General Monte Carlo N-Particle Transport Code (Version 4C), Los Alamos National Laboratory, LA-13709-M.

3. Currie, L. A. (1968). "Limits for Qualitative Detection and Quantitative Determination, Application to Radiochemistry", Analytical Chemistry 40(3):586-593.