MEANINGFUL CHARACTERISATION OF HIGH BETA GAMMA RADWASTE BY RADIOMETRIC INSTRUMENTATION

Alan P Simpson

BNFL Instruments Ltd., Pelham House, Calderbridge, Cumbria CA20 1DB, UK
Tel: +44 (0) 19467-85231 Fax: +44 (0) 19467-85203

ABSTRACT

A diverse range of wastes are generated from reprocessing spent light water reactor (LWR), advanced gas cooled reactor (AGR) and magnesium alloy (Magnox) clad uranium fuels. Much of this waste has a high beta gamma activity due to the presence of activation products and residues of spent fuel. As the processes for treatment and storage of the waste have evolved, so has the radiometric instrumentation used to monitor it. The requirements of national regulators and repository operators are becoming increasingly stringent in terms of the degree of characterisation of waste products prior to ultimate disposal. BNFL Instruments Ltd has developed several generations of radiometric instrumentation to produce accurate and reliable measurements on current arisings of the waste as it is generated. These systems use high integrity software to generate a valid inventory for individual waste products as well as providing useful process control information and ensuring criticality safety. Similar measurement systems are also required for the characterisation of historic wastes which will soon be retrieved from storage facilities. These measurements present a greater challenge due to the highly variable nature of the materials and the length of storage (up to 40 years). Solutions have been developed based on the operational experience gained from process measurements. All systems are designed such that they are capable of providing fully reproducible and meaningful measurements within a plant environment.

INTRODUCTION

The early history of nuclear power in the UK was dominated by the need to maximise the utilisation of the first generation Magnox reactors. Spent fuel was reprocessed at Sellafield, and the plants on this site were under particular pressure to run with minimal disruptions. This was the background against which the first arisings of radioactive waste were generated and stored.

Later, as the industry matured, more detailed records of the stored waste were required. Characterisation of newly generated waste using radiometric instrumentation enabled plant operators to monitor the efficiency of their processes as well as providing a means of validating the records.

Today, all operations in the nuclear industry comply to strict safety standards laid down by national regulators. Waste management procedures must protect plant operators, the public and the environment and great care is taken to ensure that waste is dealt with safely. The trend towards ultimate deep disposal of waste has also led to increasingly detailed requirements for characterisation of the waste. Physical and chemical properties of the waste and the radionuclide contents of individual waste packages must be recorded. This applies both to freshly generated waste and the historic waste which will be retrieved from storage and repackaged for disposal.

EARLY DAYS

The Magnox reprocessing plants at Sellafield have been receiving fuel from UK and foreign reactors since the 1950s. After removal from the reactor site, the fuel is shipped to the Sellafield storage ponds. Here, after a minimum period of cooling to allow short lived fission products to decay, the fuel is unloaded from the pond and sent for reprocessing. The first stage is the removal of the outer Magnox cladding (a process known as decanning). This has led to the production of significant quantities of waste mainly composed of pieces of Magnox metal of various sizes contaminated with irradiated fuel. Originally, this material (known as swarf) was dispatched to interim storage facilities.

These facilities, whilst designed according to the safety standards of the time, were relatively unsophisticated by today's rigorous standards. Concrete silos were built above ground level and filled with swarf and other radwaste arisings from Sellafield. The silos were divided into compartments to improve filling efficiency.

The first silo began receiving waste in 1952 and had reached capacity after twelve years. Here waste was stored under dry conditions. In contrast, the second silo to be built was water filled. Between 1965 and 1990 some 10,000 m3 of swarf and other waste has accumulated in this facility. Since 1990 fresh arisings of Magnox swarf have been directly encapsulated into 500 litre drums ready for ultimate disposal.

THE FIRST WASTE MEASUREMENTS

The sentencing of swarf into the dry silo and the first six compartments of the wet silo was performed without any on-line radiometric measurements. In the early 1970s, a requirement was established for the non-destructive measurement of swarf before it was emptied into the silo in order to quantify the uranium content of each bin of waste. This enabled the operators to monitor the efficiency of the decanning process. In addition the measurement would assist in fuel accountancy.

The system that was developed to meet this measurement need was based on low resolution gamma spectrometry. The emissions from irradiated fuel (up to 4 years cooling) include a distinctive high energy gamma ray at 2.18 MeV from ^{144}Pr. The activity of this nuclide was correlated to the uranium content using known relationships and the irradiation history of the fuel batch. This was originally based on sampling of the fuel dissolution process but later the reactor operator's declared data was used.

Two NaI(Tl) scintillation detectors were positioned on either side of the swarf bin. The bin was scanned past the detectors before and after emptying. The system was calibrated with standard fuel rods (of known irradiation history and cooling time) placed in a supporting frame in a water filled bin.

Although, mechanically speaking, this was a relatively simple system, rigorous technical assessments were carried out on the data collected during calibration, commissioning and operations. These assessments provided a vital role in ensuring that the measurement, although subject to various systematic and statistical uncertainties, could be meaningfully related to the actual fuel content of the waste bin.

SECOND AND THIRD GENERATION SYSTEMS

Monitoring of swarf at the silo continued until the new Magnox Fuel Handling Plant (FHP) came on-line in 1985. The operators of this plant required instrumentation at the point of decanning so that the despatch of swarf to the silo could be prevented if the residual uranium content of the swarf was above the flowsheet value and thus any large pieces of fuel could be removed from the swarf for reprocessing.

Measurement of the swarf's fuel content was achieved using high resolution gamma spectrometry (HRGS). Additional on-line measurement systems were also required in the decanning cell including a cooling time monitor to ensure that all fuel rods had sufficient cooling to be safely reprocessed.

In 1990 the silo was taken out of service and swarf from FHP was sent for direct cement encapsulation in 500 litre drums. This new waste product was designed to meet rigorous quality standards for ultimate disposal in order to satisfy the licensing and regulatory bodies. An additional measurement requirement was therefore established for the determination of a radionuclide inventory of the swarf. This led to the need for a third generation system to provide this inventory in addition to the measurement of residual uranium.

Fig. 1.

The resulting Swarf Inventory Monitor is illustrated in Figure 1 (together with the upstream cooling time monitor). For each batch of swarf, a HPGe detector views the waste and acquires a high resolution gamma ray spectrum. The spectrum is analysed and corrections are applied for the detection efficiency, the background from the measurement tray and self-attenuation in pieces of uranium. This provides an accurate determination of the activity of gamma emitting nuclides.

In addition to the direct measurement of these nuclides, the activities of non-measurable nuclides and the uranium mass are quantified using known relationships derived from the fuel inventory code, FISPIN [Ref. 1]. Irradiation and cooling time of the fuel are calculated by the system using ratios of the measured gamma emitters. This approach ensures that the inventory is independent of the reactor operator's declared fuel history data.

System Design

In the design of the system, high precision engineering is required to ensure that the system operates reliably with known geometry and that the validity of the measurements can be guaranteed at all times. The HPGe detector is mounted on the cell roof on a movable table and views the waste tray through a gamma ray collimator. The positioning of the detector is carefully monitored with an infra-red proximity sensor in order to ensure that all measurements are carried out in an identical geometry to the original calibration.

The electronics give ultra high count rate capability with an input rate of up to 300,000 counts per second. This is essential in order to cope with the wide dynamic range in the activity of the swarf. Special mounting and screening is used on the detector and its electronics to overcome electrical and mechanical noise. The system's fully automated operation features a number of self-checks including use of a standard radioactive source to check the quality of the gamma spectrum (including energy calibration and detection efficiency). This ensures that the detector continues to operate within specified parameters.

The instrument has been fully operational for several years and provides a reliable determination of uranium mass and a comprehensive radionuclide inventory. Detailed assessment work was performed after commissioning of the instrument in order to identify and eliminate potential biases. This has enabled the system's process parameters to be finely tuned to the actual measurement conditions that have been found to arise in the plant. This is of benefit to the plant operators as it reduces the potential for the system to overestimate the fuel content of a tray of swarf.

FISSILE MATERIAL ASSAY - TECHNIQUES

In addition to the requirement for radionuclide inventory and uranium carry-over in Magnox waste, many plant operators require an accurate measurement of the fissile nuclides in their waste product (mainly for criticality related safety). The most sensitive and robust methods for quantification of fissile nuclides in such material are based on neutron rather than gamma techniques

Passive Neutron Measurements

Passive neutron counting involves measuring the intrinsic fast neutron emission from the waste. Assay systems can be based on the detection of either total or coincident neutron emission. The latter may involve the detection of two time correlated neutrons (referred to as Passive Neutron Coincidence Counting PNCC) or multiple time correlated neutrons (Multiplicity Counting). Coincidence techniques allow the neutron signal from spontaneous fission of even isotopes of plutonium to be isolated from the ($\propto$,n) neutron signal (caused by the interactions of alpha particles with light elements such as oxygen). This is necessary when the chemical composition of the waste is poorly characterised such that the ratio of the ($\propto$,n) to the total neutron emission can vary.

Passive neutron measurements are often used to quantify the total plutonium content of alpha wastes (for example wastes produced from separation and handling of plutonium). This requires the isotopic composition of the material to be accurately known, either from HRGS or operator declared data.

Active Neutron Measurements

A direct technique for measuring the total fissile content of a waste package is active neutron interrogation. Neutrons from an interrogating source are introduced into a measurement chamber made up of moderating and shielding materials. Fast neutrons quickly slow down in the chamber by multiple elastic scattering in the moderating materials. In addition some moderation and absorption usually takes place in the measurement sample; the magnitude of which will depend on the waste matrix composition. The neutrons induce fission events in any fissile material present giving rise to the emission of secondary fast neutrons and gamma rays. It is this secondary radiation that is detected to give a direct measure of the mass of fissile material. Various techniques are used to maximise the sensitivity to the secondary neutrons while minimising the signal from the interrogating source

One widely used method of active neutron interrogation on waste is the differential die-away (DDA) technique. Short pulses of fast neutrons from a neutron generator are injected into the measurement chamber. This gives rise to a thermal neutron flux which persists for a few milliseconds. Fast neutrons arising from the induced fission events are then counted using fast neutron detector packages embedded in the chamber walls.

FISSILE MATERIAL ASSAY - MEASUREMENT SYSTEMS

The DDA neutron interrogation technique has been successfully deployed on Sellafield plants for the measurement of bulk quantities of high beta gamma radwastes (including Magnox and oxide fuel residues). Two examples are the Fissile Material Detector and the Thorp Hulls Monitor.

The Fissile Material Detector

The first on-plant application of DDA at Sellafield was the Fissile Material Detector (FMD) at the Miscellaneous Beta Gamma Waste Store (MBGWS). This store receives general items of high activity waste from the reprocessing plants. These items are repackaged and stored in $3m^3$ boxes. The FMD measures the fissile content of the waste for criticality safety purposes prior to filling these boxes.

The system is capable of measuring the wide variety of wastes consigned to the store. Various calibrations were performed for each declared waste classification during commissioning including mixtures of steel, lead, concrete, graphite, cellulose and plastics. The total fissile content is derived using the DDA measurement signal, operator declared classification and measured neutronic properties of the waste consignment.

The measurement chamber consists of lead, polyethylene and graphite, with neutron detectors and a pulsed neutron generator located in the walls. Automated checks on performance are carried out using a source transfer system.

The Thorp Hulls Monitor

Oxide fuel reprocessing at the THORP plant is another source of high beta gamma radwaste. Residues of fuel assemblies (known as hulls) are left behind in the dissolver after the shear/leach process. Before the hulls can be exported to the encapsulation plant they must be monitored to satisfy the requirements outlined in Table I

The measurement is undertaken on the dissolver basket (0.67m in diameter) filled with hulls up to a depth of 2m. In addition, the basket will contain fuel assembly hardware (such as end appendages) and a small amount of neutron poisoned dissolver liquor trapped within the hulls. The design of a system to meet the stringent measurement requirements under these conditions within a highly active operating cave environment represented a major challenge.

The solution was a combination of DDA, passive neutron totals counting and HRGS. Figure 2 illustrates how the measured data is combined with fuel data (cooling time and initial enrichment) and FISPIN derived correlations to provide the required parameters. Previous experience in these techniques described above was an essential foundation for designing a reliable fully automated system. The resulting Hulls Monitor is illustrated in Figure 3.

Fig. 2.

Fig. 3.

When the operator initiates a measurement sequence, the basket is lowered into a re-entrant thimble in a monitoring cell below the dissolver basket handling area. Fast neutron detectors and the neutron generator are housed in a collar constructed of moderating and shielding materials which surrounds the thimble. The collar acts as the thermalisation ‘chamber’ for the DDA measurement. A separate HRGS system is located outside the cell and views the basket through a collimator set into the cell wall above the neutron collar.

Many features of the instrument were designed to cope with the measurement environment. Lead shielding is used to reduce the high gamma flux at the neutron detectors. Plant ruggedised electronics were employed to provide the detection systems with noise immunity, high count rate capabilities and fast recovery times essential for DDA measurements. High integrity software is used to supervise instrument operations, acquire and process measurement data and to manage the on-line interfaces to plant computer systems.

To provide a high level of reliability, comprehensive self-checking and back-up facilities were designed into the instrument. Functionality of the detection systems and the neutron generator is confirmed by automated standardisation checks. These are initiated by the basket handling cave control system at regular intervals and before each measurement. Fail-safe features are in place to prevent a measurement being carried out without confirmation of a satisfactory standardisation. In addition, continuous real-time checks are performed by the software to confirm the absence of fault conditions during the measurement period.

The hulls monitor provides a reliable measure of the residual fissile content of a basket of hulls. The system calibration was set to ensure that the measurement result represents a maximum fissile content in order to satisfy the criticality safety case. Although this measurement is designed primarily as a guard against high fissile loadings in the basket (of the orders of several hundreds of grams), the lower limit of detection of the instrument is around 5-10 g ^{235}U equivalent depending on the waste matrix. The sensitivity of the system provides the operators with a high level of confidence in the measurement result.

WASTE RETRIEVAL MEASUREMENTS

The next generation of radiometric instrumentation on the Sellafield site will be used in the assay of materials retrieved from interim storage silos (described above) and redundant fuel storage ponds. These waste streams will include mixtures of swarf, sludges, corroded fuel and miscellaneous items of scrap including metals, organics, graphite and concrete. The retrieval, repackaging and encapsulation of this waste is part of BNFL’s overall waste management strategy [Ref. 2]. When these facilities are emptied it will be possible to complete the decommissioning process by dismantling the building itself.

Characterisation Requirements

The post-operational clean out (POCO) of the ponds and silos will generate waste packages which require characterisation on an individual basis. In-situ sampling and plant records can often provide a good representation of the overall contents of the waste stores. However, the characterisation requirements for individual items of waste product cannot be easily satisfied with this information alone. As with current arisings of waste, one of the major requirements is the provision of radionuclide inventory and fissile content information to satisfy disposal regulations and to ensure criticality safety.

Waste Measurement Conditions

Typically, waste is retrieved in batches, which contain a variable mixture of materials. Treatment, sorting and repackaging are often performed before the monitoring stage. These measurement conditions are also often complicated by the length and method of storage, the inhomogeneous and variable nature of the materials and the use of large volume waste containers. Further problems arise due to the shielding, handling and containment requirements dictated by high dose rates and loose contamination associated with the waste treatment processes.

Technique Selection

Providing a technique for the accurate assay of historic wastes presents a major challenge. Gamma measurements alone are of limited applicability for the measurement of large drums of historic waste for the following reasons:
- The short half-lives of many of the gamma emitters;
- Interference effects from the intense gamma rays from fission and activation products;
- Absorption of gamma rays in bulk materials (particularly severe for scrap metals);
- Prolonged contact with water (during wet storage) leads to selective leaching of fission products which adversely affects the ability to use isotopic ratio fingerprints.

Passive and active neutron techniques are better suited to the measurement of these wastes. However, there are a number of problem areas in the adaptation of these techniques to the highly active wastes retrieved from interim storage. Some of the challenges that have been encountered and the solutions developed are summarised in Table II. The fully developed assay system can operate with sensitivities at or below gram levels of fissile material.

ADVANCED MEASUREMENT SYSTEMS

Further work is underway to develop improved systems for the measurement of more challenging high beta gamma waste streams. Current areas of development include:
- The measurement of hulls from mixed oxide and higher irradiation uranium oxide fuels. High passive neutron emissions for these fuels lead to the requirement for a more intense interrogating neutron source than conventional D-T tubes.
- The measurement of waste within a concrete (encapsulation) matrix. This leads to severe absorption of passive emissions and most forms of interrogating radiation.
- Instrumentation to aid the waste sorting and treatment process as well as characterisation. Gamma imaging techniques are being developed for this purpose.

CONCLUSIONS

Several generations of radiometric instrumentation have been developed for the characterisation of current arisings of high beta gamma radwaste prior to storage and ultimate disposal. Successive systems have become more advanced to meet the increasingly detailed requirements of plant operators, national regulators and disposal bodies.

These computer controlled instruments must perform reliable on-line measurements within challenging plant environments. Shielding, handling and containment are necessary due to high dose rates and loose contamination. Measurement of historic waste retrieved from storage presents further difficulties due to the age of the material and its highly variable nature.

Operational experience has been applied to all areas of technique development and system design (including the electronics, hardware and software). This leads to systems which are able to operate reliably with a wide variety of waste types. This experience combined with a quality based approach in the design, commissioning and operation of characterisation systems, provides operators and regulators with confidence that the measured result represents an accurate record of the fissile and radionuclide content of the waste.

REFERENCES

1. R.F. BURSTALL, “FISPIN - A Computer Code for Nuclide Inventory Calculations,” ND-/R/328(R), United Kingdom Atomic Energy Authority (October 1979).

2. A.D. POWER and P.E. VICKERY, "Intermediate Level Waste Management at Sellafield - A Comprehensive Strategy," Proc. Int. Conf. Radioactive Waste Management and Environmental Remediation, Berlin, Germany, (September 3-7, 1995).

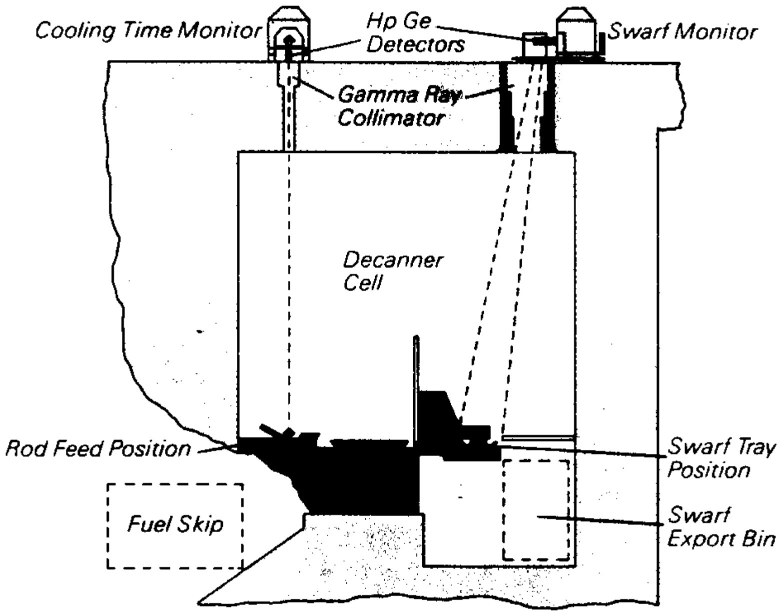

Fig. 1 Mechanical Arrangement of the Swarf Inventory Monitor within the Decanning Cell

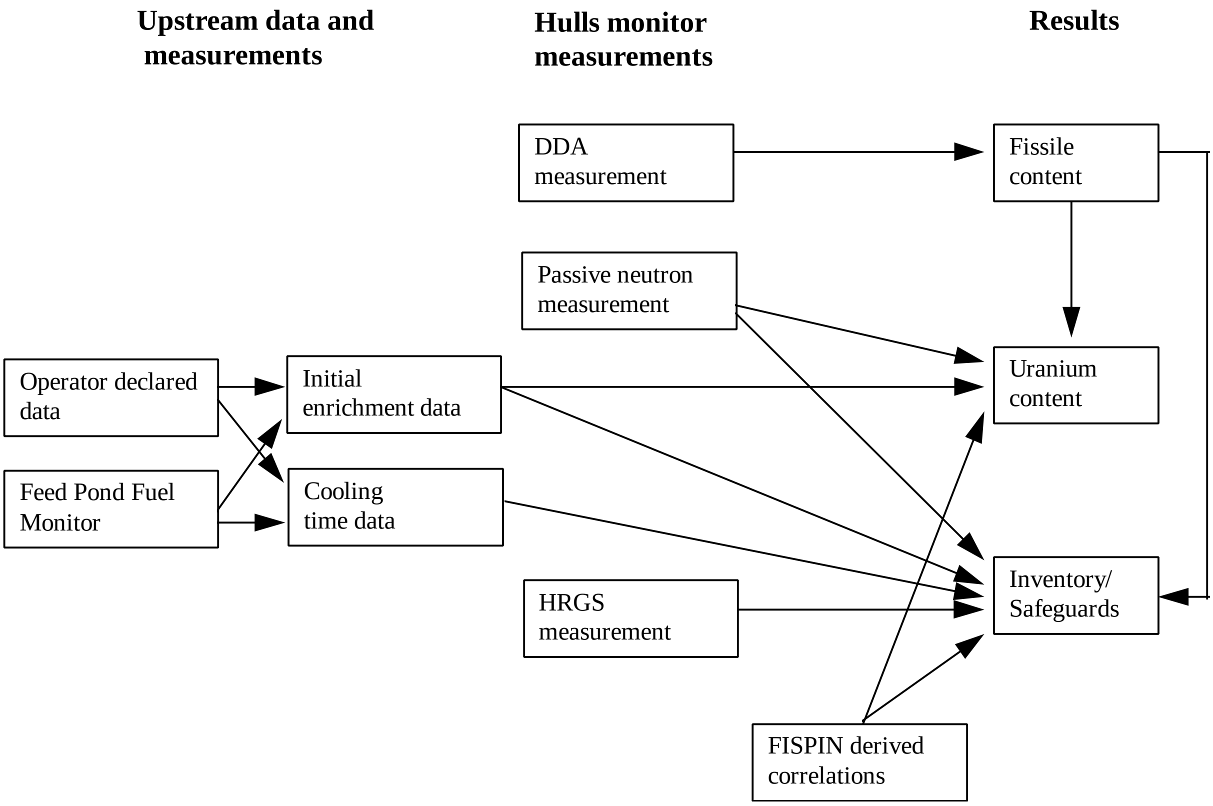

Fig. 2 Information Flow in the THORP Hulls Monitor

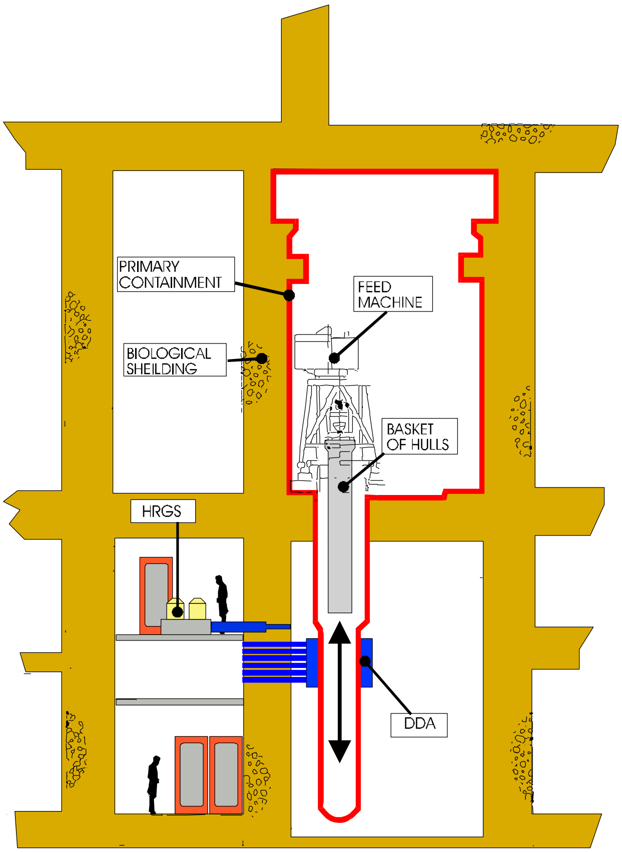

Fig. 3 Schematic of THORP Hulls Monitor

TABLE I
Monitoring Requirements For Hulls Prior to Export

Monitoring Requirement	Parameters to be Measured
Ensure criticality safety in subsequent handling	Fissile content
Ensure that fuel retention complies with limits for interim storage and ultimate disposal or return to the customer	Total uranium content. ^{60}Co, ^{137}Cs, ^{134}Cs, ^{106}Ru activities. Total alpha, total plutonium, total uranium, ^{244}Cm activity
Demonstrate that uneconomic fuel retention has not occurred	Total uranium content
Derive activity inventory information for customers and regulators	Inventory of isotope activities
Provide safeguards and materials accountancy data	Fissile content, fissile uranium, total uranium, fissile plutonium, total plutonium

TABLE II
Challenges in the Application of Neutron Based Techniques for Measurement of Retrieved Wastes

Problem	Description	Solution
Variable composition	Materials that have very different neutronic properties (absorbers and moderators) are often found together. Examples include graphite, stainless steel, organics and sludges	Optimum performance is achieved by segregation of materials by the operator. Measurement based matrix correction techniques can then be used to compensate for smaller compositional variations within each waste category
Water content	For neutron measurements, the moderating effect of water is usually undesirable, particularly where the water content is variable	Efforts should be made to present the waste in a dry state. Experience shows that small quantities of water will not cause a significant problem
Variable density	Typical bulk density ranges that can be encountered are 0.5 to 5 g/cm^3. This variation can have a large effect on absorption and moderation properties of the waste	Weighing of the waste container enables the bulk density to be determined. Monte Carlo calculations can be used to extend the system calibration to deal with variations in bulk density
Large containers	Large containers are undesirable from a measurement point of view as the increased degree of neutron absorption leads to high measurement uncertainty	Efforts should be made to measure the waste in as small a package as possible prior to filling any larger container An imaging capability has been developed to correct for variations from a uniform distribution of fuel.