Difficult But Not Impossible: Monitoring Highly Active Wastes Retrieved From Interim Storage Facilities

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ABSTRACT

Major national nuclear programmes have resulted in a considerable number of storage facilities containing either highly active liquors or highly active solid wastes. The latter in many cases contain inhomogeneous mixtures of fuel, metal scrap and debris. In the UK major efforts are underway to retrieve these wastes for treatment and, ultimately, disposal. In order to conduct these operations safely, and to meet the inventory data requirements for ultimate disposal, it is essential to achieve a valid measurement of inventory in the early stages of the waste routing process. BNFL Instruments Ltd are playing an important role by providing this measurement capability to retrieval projects on the parent company's Sellafield site. Current measurement solutions are based on many years of operational experience which the company has accumulated in delivering special purpose waste monitoring instrumentation for process control as well as for inventory measurements. In spite of the difficulties of measuring the inventories of very variable retrieved wastes, in high dose rate environments, measurement systems are being provided which satisfy the demands of both the repository operators and national regulators.

INTRODUCTION

In common with the other nuclear powers, Britain has constructed and operated several generations of nuclear reprocessing facilities since the early 1950's. The Sellafield site, now operated by British Nuclear Fuels plc (BNFL), has seen the construction and operation of early fuel storage, reprocessing, product finishing and waste storage facilities which are now in varying stages of shutdown or decommissioning. Of particular concern are the very variable solid wastes which may be contaminated with a variety of fissile and radioactive nuclides. Interim storage facilities within the site boundary have contained these highly active wastes safely and with minimum environmental impact for several decades. Now as part of BNFL's overall waste management strategy, major efforts are underway to empty these stores and repackage and encapsulate the waste prior to ultimate disposal. Once emptied the buildings themselves can be decommissioned.

In this paper the issue of measurement is placed in the context of the waste retrieval operations and the particular difficulties of adequately monitoring the radionuclide inventory and fissile content are highlighted. The solution to such problems can be shown to lie in the experience gained by close involvement throughout more recent reprocessing and waste treatment operations. BNFL Instruments Ltd, a wholly owned subsidiary company of BNFL plc, have supported the parent company's engineers and designers by developing specialised instrumentation for all new plants at Sellafield for over 25 years. In the course of this support, techniques have been developed which, for highly active waste, will enable operators to perform a non-destructive assay prior to encapsulation and to establish the detailed characterisation of waste required to satisfy the disposal authority and the national regulator.

WASTE STORAGE AND RETRIEVAL

An overview of the material flow in reprocessing operations and the routes by which wastes arise at Sellafield are shown in Figure 1. Irradiated fuel has historically been stored in water filled ponds prior to reprocessing. Once sufficiently cooled for safe reprocessing, the fuel is taken for pre-treatment. For Magnox fuel, this consists of stripping off the outer cladding prior to dissolving the metal fuel in nitric acid. The de-cladding debris (known as swarf) is treated as intermediate level waste (ILW). Current arisings of swarf are being sent directly for cement encapsulation into 500 litre drums, at a plant commissioned in 1990. A second plant commissioned in 1992 is used to deal with the cladding residue from oxide fuel reprocessing in a similar way.

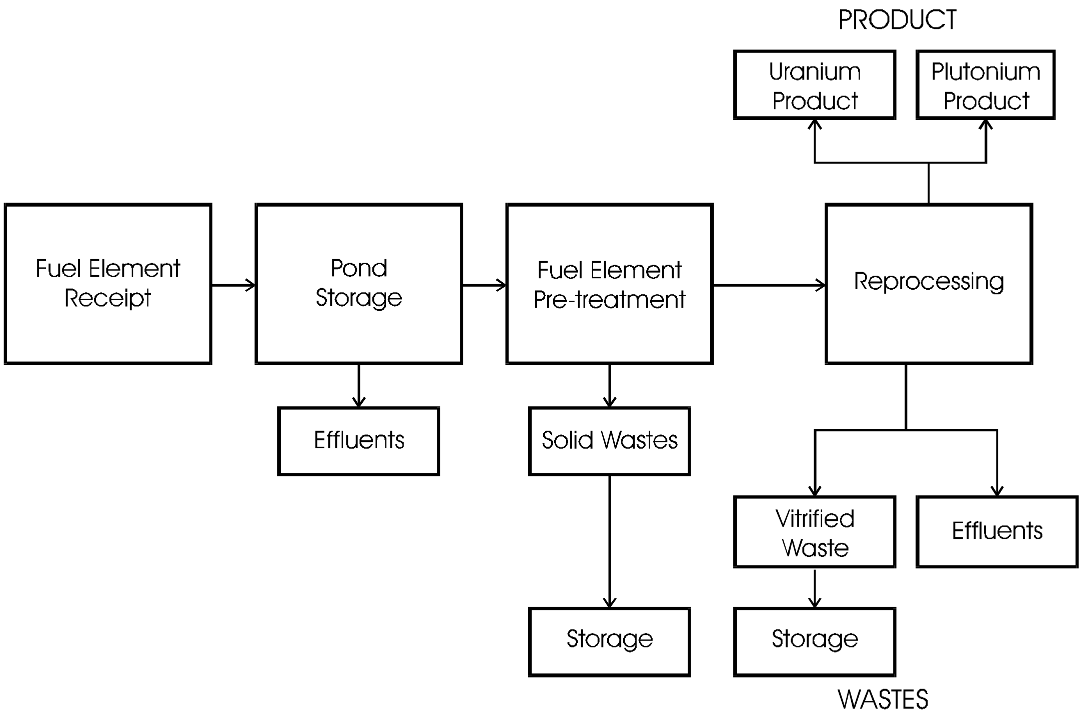

Fig. 1
Reprocessing Flowsheet

Storage

Prior to the availability of the encapsulation plants, the cladding waste was sent to interim storage facilities. The first such facility was a dry silo, but from 1960 onwards new arisings were stored underwater. The silos were also often used to store scrap associated with the de-cladding process. The contents of the silo compartments filled in the early years of operation have corroded to a mixture of sludges and solids. The original fuel storage ponds, which are now redundant, also contain sludges, corroded fuel and fuel handling equipment.

Retrieval

As part of BNFL's overall waste management strategy, major efforts are underway to empty the redundant ponds and interim storage facilities and repackage and encapsulate the waste [Ref. 1]. Once emptied it will be possible to complete the decommissioning process by dismantling the building itself.

Safety and dose minimisation are important issues in engineering a cost-effective retrieval process. Major problems to be overcome include: high dose environments, the need for containment to prevent the spread of contamination, chemical hazards, fire risks and other safety issues. Significant development work and plant trials have been undertaken to find the safest and most cost-effective means of retrieval. Purpose built machinery is being designed and constructed to facilitate retrieval and subsequent treatment of the stored waste into a form suitable for ultimate disposal. Several dedicated plants will be built to receive this waste. Sorting, screening, settling, packaging and drying prior to encapsulation of the waste are all options either in operation or under consideration.

The retrieval of stored irradiated fuel bearing wastes has already begun on the Sellafield site and will continue well into the next century.

Measurement and characterisation

Detailed reviews including sampling, analysis and scrutiny of plant records have been carried out on the contents of the redundant storage facilities. This has enabled sufficient information on the stored materials to be determined for the selection of waste routing through to disposal.

The waste retrieval, treatment and repackaging process will generate waste packages which will require characterisation on an individual basis. The sampling and plant records can often provide a good representation of the overall contents of the waste stores, however, the characterisation requirements for the final waste product cannot be satisfied with this information alone. One of the major requirements is the provision of radionuclide inventory and fissile content information to satisfy disposal regulations and to ensure criticality safety.

Non-destructive assay (NDA) techniques can be used for measurement of each waste package prior to ultimate disposal. The requirements for NDA depend on the application. There are a number of general driving forces including safety, dose minimisation, regulatory requirements, disposal authority requirements and process control.

The major function of waste product monitoring is the generation of an extensive radionuclide inventory including fission products, trans-uranic radionuclides and total fissile content of the waste, where applicable.

CHALLENGES FOR WASTE MEASUREMENT

In determining the optimum monitoring solution, physicists, engineers and designers have co-operated to find the most suitable waste retrieval strategy incorporating the required measurements. The technique used, location and physical arrangement of the system must fit in with the need to engineer a safe and cost-effective waste routing process through to disposal, taking into account the suitability of measurement at various possible locations.

Typically, waste is retrieved in batches, which contain a variable mixture of materials including highly active fuel residues, fuel cladding materials and miscellaneous operational scrap. Figure 2 illustrates a mock-up of typical retrieved wastes undergoing a sorting process. Treatment, sorting and repackaging are often performed before the monitoring stage. The detailed inventories that have been prepared for the storage facilities themselves will generally provide only limited information for the waste measurement system to use (e.g. during calibration). These measurement conditions are also often complicated by the bulk quantities of waste involved and the inhomogeneous and variable nature of the materials.

Fig. 2
Typical Retrieved Wastes

Further problems arise due to the high dose rates and potential for loose contamination associated with the waste treatment processes. This often leads to shielding, handling and containment requirements which makes the measurement more problematic.

Quality Assurance plays a key role throughout the entire process of providing plant operators with the solution to their measurement needs. The instrumentation must be capable of generating fully reproducible results and must operate reliably and be capable of performing regular automated self checks.

MEASUREMENT TECHNIQUES

There are a variety of radiometric NDA techniques which can be used in waste monitoring. The basis of all such measurements is the quantification of certain properties of the sample based on the detection of some form of radiation. Due to the bulk quantities of materials encountered, the most suitable NDA techniques are based on either gamma or neutron measurements.

Gamma Ray Measurements

In order to obtain the maximum amount of information from a gamma ray measurement, High Resolution Gamma Spectrometry (HRGS), based on high purity germanium (HPGe) semiconductors, is preferred to low resolution spectrometry based on scintillation detectors. Within irradiated fuel, there are many gamma emitting nuclides. Due to the attenuation effects within the waste container, only higher energy gamma rays can be measured in large containers. In practice there are very few photopeaks in the spectrum of historic irradiated fuel bearing waste. This is mainly due to the long cooling times and Compton scatter interference effects from several high energy, high intensity gamma rays (mainly ^{137}Cs and ^{60}Co).

Gamma ray detection can also be used by instrumentation at the retrieval facility and within the treatment and sorting facilities. Gamma ray imaging techniques have been developed which allow operators to identify ‘hot spots’ of gamma activity on a contaminated wall, say, or at a sorting table. These instruments can include a coarse spectrometric capability which enables fuel to be discriminated from other items. This ‘gamma view’ can be combined with the traditional optical image to assist in segregation of the waste and reduces operator dose uptake.

Passive Neutron Measurements

Passive neutron counting involves measuring the intrinsic fast neutron emission from the waste. This arises from two types of event: spontaneous fission and (∝,n) reactions. Typically 2 or 3 coincident neutrons will be emitted from each spontaneous fission event, whilst (∝,n) reactions (caused by the interactions of alpha particles with light elements such as oxygen) result in the emission of a single neutron. Maximum (∝,n) emission occurs when the alpha emitter is chemically linked to the light element as, for example, in corroded fuel.

Assay systems can utilise the detection of the 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 signal from spontaneous fission to be isolated from the (∝,n) signal. This is necessary when the chemical composition of the waste is poorly characterised such that the ratio of the (∝,n) to the total neutron emission can vary.

Active Neutron Interrogation

In contrast to passive methods, active neutron measurements rely on the detection of induced radiation. 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 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 measurement of the total amount of fissile material present.

One method of active neutron interrogation 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. These detector packages have much shorter characteristic neutron lifetimes than the chamber and this large difference in the die-away time makes the measurement possible. The measurement signal is used to quantify the mass of fissile material present.

EXPERIENCE IN WASTE MEASUREMENTS

Over the last 25 years, many special purpose waste monitoring systems have been developed to meet a wide variety of difficult measurement problems in the Sellafield reprocessing and waste treatment operations [Ref. 2]. As new generations of increasingly sophisticated plant have been commissioned and the demands of the regulators have become more stringent, several generations of instrumentation have been developed from concept through to commissioning, calibration and fully automated operation. Several examples are given here of such systems.

Swarf Tipping Monitors

One of the earliest measurements of highly active wastes was performed during the process of transferring the Magnox decanning waste to its interim store. A measurement of the uranium content of the waste was required in order to quantify carry-over of fuel from the decanning plant. The chosen measurement technique was based on detection of a distinctive high energy gamma ray from a short cooled gamma emitter which is only associated with irradiated fuel (the 2.18 MeV gamma ray from ^{144}Pr was used).

Two low resolution NaI(Tl) gamma spectrometers viewed a swarf bin which was scanned past the detectors before and after tipping. Calibration was performed using fuel rods of known irradiation history and cooling time, placed in a supporting frame, at known positions in a bin full of water.

The measurement enabled the reprocessing plant operators to gain confidence in the decanning process and was also of sufficient quality to contribute to the materials accountancy for the plant. A great deal of performance assessment work was carried out during and after commissioning of the instrument. This work has provided confidence in the accuracy of the declared data and provided valuable experience in the development of later generations of highly active waste assay instrumentation.

Swarf Inventory Monitor

The next generation of monitoring instrumentation was developed to improve on the original measurements carried out during swarf tipping. In 1985, the first high technology system (designed to quantify fuel carry-over), was installed in a new fuel handling plant. Subsequently a system was developed to provide improved measurements in parallel with the advent of swarf encapsulation. As previously noted, since 1990 arisings of waste from the Magnox decanning process have been encapsulated into 500 litre drums to await ultimate disposal. A requirement of the licensing and regulatory bodies is that an extensive radionuclide inventory must be provided for this waste. In addition, there is a process control measurement requirement, to monitor for excessive quantities of uranium in the swarf prior to export to the encapsulation plant. The most appropriate measurement technique is HRGS, since the relatively short cooling time of this waste stream enables a wide variety of nuclides to be directly measured. This provides sufficient information to determine the irradiation history of the fuel from which it is possible to infer the uranium content as well as a variety of other non-measurable radionuclides.

Measurements on bulk quantities of swarf would be undesirable because of gamma absorption effects, so the swarf is measured in small batches. The Swarf Inventory Monitor (SIM), located in the decanning cell, is illustrated in Figure 3 (a neighbouring instrument for measuring the cooling time of the fuel is also shown here). For each batch, a HPGe detector views the waste and acquires a gamma ray spectrum. The activity of the gamma emitters is calculated from analysis of the spectrum. This calculation takes into account detection efficiency, background from the measurement tray and self-attenuation in pieces of uranium. The irradiation history of the fuel is derived from various ratios of the measurable gamma emitters. Uranium mass and the activity of other radionuclides can then be quantified using known relationships derived from the fuel inventory code, FISPIN [Ref. 3].

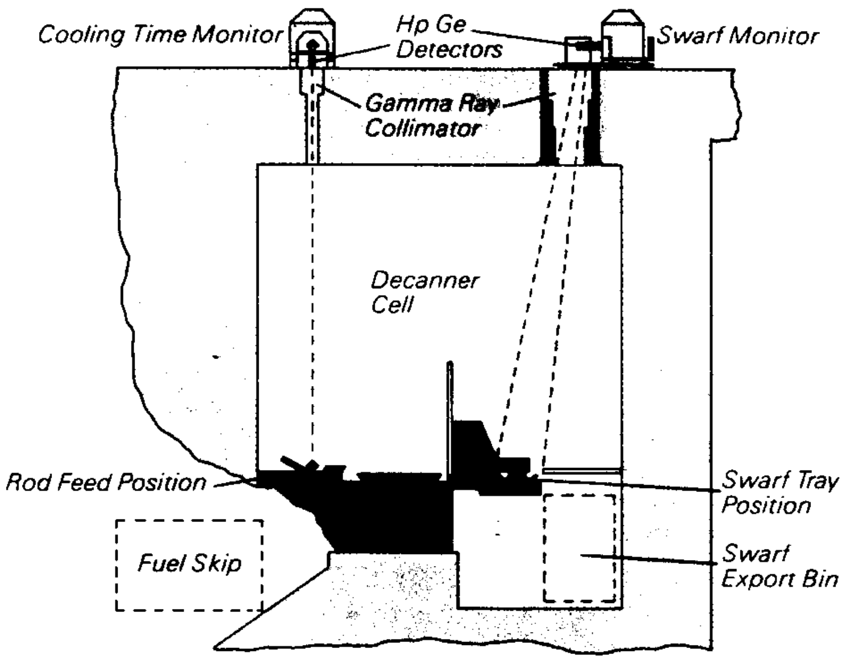

Fig. 3
Mechanical Arrangement of the Swarf Inventory Monitor

The HPGe detector is mounted in the cell roof on a precisely engineered movable table. The detector 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 the calibration arrangement remains valid during all subsequent measurements. An ultra high count rate capability (handling an input of up to 500,000 counts per second) is required to provide a wide dynamic range to cope with the variation in activity of monitored swarf. Advanced electronics are used to process the signal from the detector with accurate dead-time correction. Based on experience gained in using HPGe detectors in process plants, special mounting and screening is used on the detector and its electronics to overcome electrical and mechanical noise. Considerable effort was made to provide high quality diagnostics and to make the system 'user friendly' for the plant operators and engineers.

The monitor has been operating since July 1990 and provides a reliable determination of uranium mass and 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.

Fissile Material Detector

Current arisings of miscellaneous items of ILW from the Sellafield plant are stored in $3m^3$ boxes at a purpose built facility, the Miscellaneous Beta Gamma Waste Store (MBGWS). For criticality safety, it is necessary to quantify the fissile content of this waste prior to filling of the storage boxes. The technique employed for this measurement is the active neutron interrogation technique, DDA.

The Fissile Material Detector (FMD) is capable of measuring the wide variety of wastes consigned to the store. Various calibrations were performed for each type of waste classification during commissioning. The waste consigned to the plant includes miscellaneuous mixtures of: steel, lead, concrete, graphite, cellulose and plastics. The total fissile content is derived using the DDA measurement result, operator declared classification and the neutronic properties of the sample, determined during the measurement.

The measurement chamber consists of polyethylene and graphite, with neutron detectors and a pulsed neutron generator located in the walls. Lead shielding is used to reduce the gamma flux to the detectors from the waste. Regular automated checks on the system performance are performed using a source transfer system.

Hulls Monitor

The reprocessing of oxide fuel at the THORP plant generates another ILW stream. This consists primarily of the residues of fuel assemblies (“hulls”) which are the waste product from the dissolver after the shear/leach process. The hulls are exported from THORP to an encapsulation plant where they grouted and stored prior to ultimate disposal. Monitoring of the hulls (prior to export) is necessary in order to:-

- Ensure criticality safety in subsequent handling.
- Ensure that fuel retention complies with limits for interim storage and ultimate disposal or return to the customer.
- Demonstrate that uneconomic fuel retention has not occurred.
- Derive inventory information for customers and regulators.
- Provide safeguards and materials accountancy data.

The Hulls Monitor, illustrated in Figure 4, has been developed to satisfy these measurement requirements using a combination of neutron interrogation (DDA), passive neutron totals counting and HRGS. The application and development of these techniques to this measurement scenario represents a major measurement challenge.

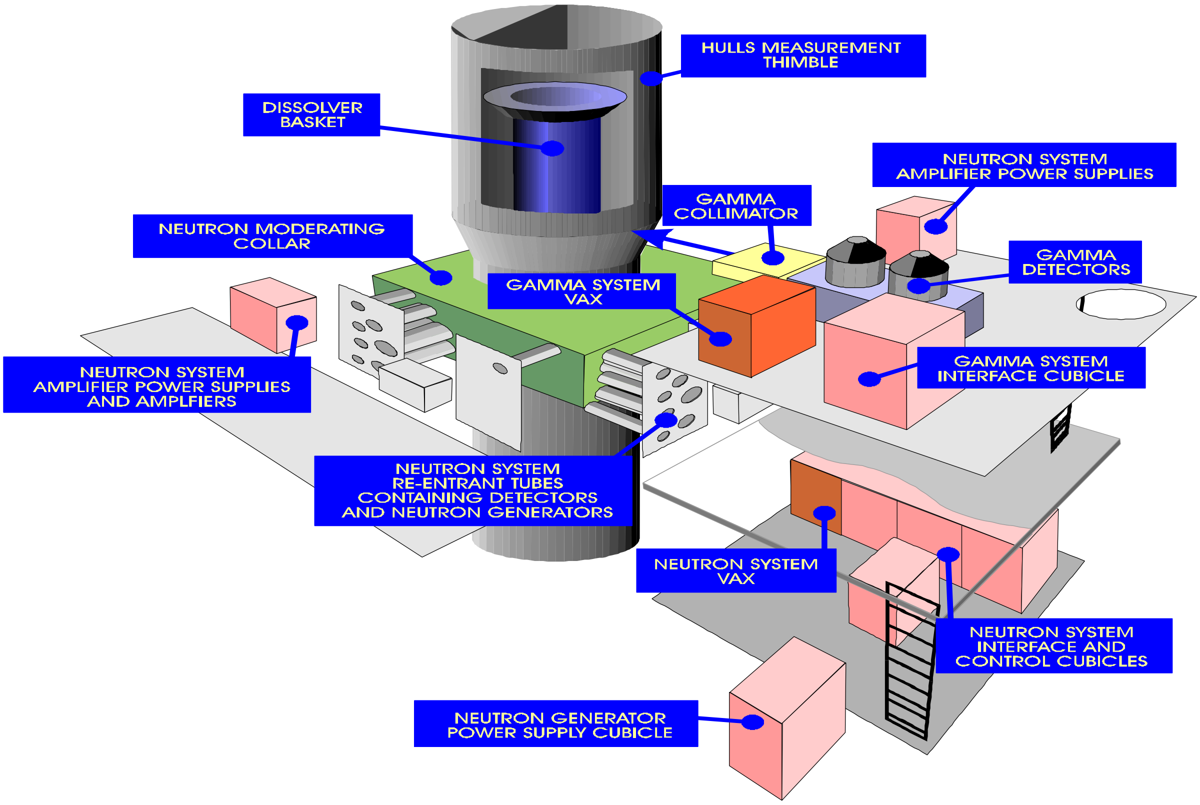

Fig. 4
Schematic of Hulls Monitor

The residual fissile content of the leached hulls is determined from the DDA measurement. Total uranium content is derived using the fissile content, the measurement of passive neutron emission and information on the initial enrichment of the fuel batch provided by the reactor operators and from measurements (via a separate monitor) on the fuel before shearing. Additional inventory information is determined using HRGS, the passive neutron count, initial enrichment and cooling time of the fuel and FISPIN derived correlations.

The measurement is undertaken on a dissolver basket, 0.67m in diameter filled with hulls up to a depth of 2m. In addition to the hulls, the basket will contain additional fuel assembly hardware (such as end appendages) and a small amount of neutron poisoned dissolver liquor trapped within the hulls. During a measurement sequence, the basket is lowered into a re-entrant thimble in a monitoring cell below the basket handling area. Fast neutron detectors and the neutron generator are housed in a collar around the thimble. The collar is constructed of moderating materials designed for the DDA measurement. Plant ruggedised electronics have been developed to provide the detection systems with noise immunity, high count rate capabilities and fast recovery times essential for DDA measurements. The HRGS system is located outside the cell and views the basket through a collimator set into the cell wall above the neutron collar.

Comprehensive self-checking and back-up facilities have been designed into the instrument. Functionality of the neutron and gamma detection systems, and of the neutron generator is confirmed by standardisation checks that are initiated by the basket handling cave control system at regular intervals and before each measurement. Confirmation of a satisfactory standardisation must be provided before a measurement can be carried out. In addition, real-time checks are continually performed by the software to confirm the absence of fault conditions.

The hulls monitor neutron system is capable of measuring residual fissile content with a lower limit of detection of 5-10 g ^{235}U equivalent levels. Further work is underway to develop an advanced radiometric instrument to deal with the more challenging measurement of hulls from mixed oxide and higher burn-up uranium oxide fuels.

THE MEASUREMENT SOLUTION

Development work has been performed to adapt the existing waste measurement technologies to the challenge of monitoring highly active waste retrieved from interim storage facilities. One area in particular where considerable development work has been applied is in the application of neutron assay techniques. These measurements are often needed for criticality control and as supporting information for radionuclide inventory of waste packages prior to ultimate disposal.

Technique development

The assay of trans-uranic elements in highly active waste presents a major measurement challenge. Passive techniques, in isolation, are of limited applicability. The gamma rays associated with the trans-uranic nuclides are often difficult to detect due to the low energy or low emission rates. For the measurement of irradiated fuel bearing wastes, there is usually no measurable gamma signal from any trans-uranic nuclide because of the interference from the intense gamma rays from fission and activation products. Passive neutron counting can be used on highly active wastes but this measurement can only quantify the total spontaneous fission neutron emission rate (due to emissions from even mass nuclides such as ^{238}U, ^{240}Pu and ^{244}Cm). In isolation, this measurement cannot be used to quantify specific trans-uranic nuclides. To do this, additional parameters relating to the trans-uranic content of the waste need to be known or measured.

Active neutron interrogation enables the total fissile content of the waste to be measured (comprising the fissile nuclides ^{235}U, ^{239}Pu and ^{241}Pu). For waste measurements, one of the most suitable forms of this technique is DDA. In the UK, as elsewhere in the world, the earliest applications of DDA have been the assay of low concentrations of fissile material in wastes that have low overall inventories of radioactive materials. In addition the materials presented for assay and the geometrical arrangements have traditionally been well characterised.

The adaptation of DDA to the highly active wastes retrieved from interim storage is complicated due to the following aspects of the measurement:-

- *Wastes can vary in composition.* Materials that have very different neutronic properties (absorbers and moderators) are often found together. Examples include graphite, stainless steel, organics and sludges.
- *Waste is heterogeneous.* Frequently a container will contain voids and lumps of different materials.
- *Water is often present in the waste.* For neutron measurements, the moderating effect of water is usually undesirable, particularly where the content is variable. Efforts can be made to design the waste retrieval process to present the waste in a dry state. However, safety or process limitations often lead to the presence of bound or free water in the waste stream.
- *Wastes can vary in density.* Typical ranges that can be encountered are 0.5 to 5 g/cm^3.
- *The wastes are poorly characterised.* A methodology has been developed for measurements of mixed wastes, where the operator cannot define distinct ‘streams’.
- *The measurement container is large.* Process and engineering requirements often mean that the waste can only be presented for measurement in large containers. This is undesirable from a measurement point of view as it

increases the uncertainty in the measurement. Physicists have worked in close collaboration with plant designers to find the most appropriate measurement point in the process.

System Description

The conceptual measurement system for assay of highly active waste comprises a custom built chamber made up of polyethylene and graphite, suitable for both active and passive measurements (DDA and PNCC). Fast neutron detector packages are located in the walls of the chamber together with the pulsed neutron generator(s) for the active assay. Due to the gamma emissions associated with highly active materials, the chamber is lined with several centimetres of lead shielding to reduce the gamma flux to the detectors (which are sensitive to both gamma rays and neutrons).

Imaging and matrix corrections capabilities have been developed which allow accurate determination of the fissile content and spontaneous fission neutron emission rate of a variety of waste streams. It is possible to use these parameters to quantify individual trans-uranic nuclides (e.g. ^{239}Pu) and fission products in the waste container. Sensitivities at or below gram levels of fissile material are achievable.

CONCLUSIONS

Characterising a retrieved waste from an interim store which may have been operational thirty or more years ago represents a challenge to the instrument supplier. For a site such as the BNFL Sellafield facility, many such retrieved waste streams will be encountered as redundant facilities are cleaned out in preparation for decommissioning, each posing its own distinct set of problems. To all of this can be added the challenge of meeting the increasingly detailed requirements of the regulator and disposal site operator in terms of the degree of characterisation required.

Detailed consideration of some of the needs for retrieved waste monitoring at Sellafield has demonstrated the value of utilising instrumentation concepts and techniques originally developed in support of reprocessing and associated operations.

BNFL Instruments Ltd with its considerable experience of providing instrumentation systems for the characterisation of spent fuel and freshly generated wastes, which have now been taken through several generations of increasing sophistication and reliability, has provided the parent company with the confidence to move forward in its waste retrieval programme with the knowledge that tractable and economic solutions can be developed for all waste streams.

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