CHOICE OF INSTRUMENTATION FOR COST–EFFECTIVE UTILITY WASTE MANAGEMENT

C H Orr
J C B Simpson
M Newell*
P A Clark

BNFL Instruments Ltd
Pelham House
Calderbridge
Cumbria
CA20 1DB
UK

*BNFL Instruments Inc
278 D P Road
Los Alamos
New Mexico 87544
USA

Abstract

Gamma based measurement techniques are well established for the non-destructive assay of gamma emitting radioisotopes in wastes arising from nuclear reactor operations and decommissioning. In this context, such measurements support compliance with the appropriate transportation and waste disposal criteria. Such techniques are also used, where regulations allow, to support the de-categorization of waste to the lowest possible level, including 'Below Regulatory Concern' or 'Unrestricted Release', in order to minimize waste disposal costs

Historically, such needs have been met through the use of High Resolution Gamma Spectrometry (HRGS) systems, 4Pi plastic scintillator systems and interpretation of dose-rate data. More recently, the use of Low Resolution Gamma Spectrometry (LRGS) has emerged as an attractive alternative for those applications in which the waste stream is relatively well characterized in terms of radionuclide and matrix composition.

This paper reviews the relative merits of all three approaches in relation to monitoring of packages, drums and boxed wastes arising from nuclear power plant operations and describes practical experience in the use of LRGS based systems for monitoring solid radioactive waste from a variety of sources .

Introduction

Characterization of the radionuclide inventory of solid waste is a key element of the overall waste management strategy for nuclear power plants. This includes operational waste arisings, as well as historic waste generation and waste arising from decommissioning operations.

This paper addresses the routes available to provide cost effective characterization of such wastes which may be in the form of packages or discrete items (such as HEPA filters) or contained in drums or boxes of various sizes. A selection of the most appropriate monitoring technique depends on a number of factors such as:

Radionuclides that need to be identified and/or quantified
Sensitivity required
Accuracy required
Waste item or container size
Waste matrix characteristics (material, density, degree of homogeneity)

In general, for waste arising from nuclear power plants the key concern is to determine the activity of fission and activation products. Consequently assay is generally through a combination of non-destructive assay using gamma based methods combined with both historical knowledge of the waste stream and laboratory analysis of representative samples. Sample analysis is used to determine ratios of those radionuclides that cannot be directly measured through by gamma spectrometry (for example Sr-90) to those radionuclides that are amenable to such gamma-based determination (for example Cs-137 and Co-60). The extent of characterisation that can be achieved through sampling and the degree of radionuclide variability within waste streams are key issues in the determination of the most appropriate non-destructive assay technique, for example:

Gross gamma measurements
Low resolution gamma spectrometry (LRGS)
High resolution gamma spectrometry (HRGS)

Sensitivity and measurement accuracy considerations relate to the ultimate destination of the waste from specific waste streams. This can range from monitoring to confirm that the waste is essentially non-active as part of a program to enable waste to be treated as below regulatory concern, through to monitoring of low and high active materials to meet disposal and transport regulations. For the latter, accurate characterization opens the possibility to re-categorize waste to the lowest level thereby achieving cost reductions through less onerous transport and disposal requirements. Such characterization may be integral with a waste sorting or decontamination program and may be used to both identify items suitable for decontamination as well as confirming that any decontamination technique has been successful.

The following sections detail the issues relating to the optimum choice of gamma based non-destructive analysis technique and is supported by a number of specific cased studies into the use of such techniques in real applications.

Radionuclide Inventory Determinations

Radionuclide inventory data for solid wastes generated during routine facility or decommissioning operations are usually provided by one [or a combination] of the following methods:

Gamma Spectrometry:
Radionuclides are identified and quantified using the detected spectrum from passive gamma emissions that are often uniquely characteristic of selected radionuclides. The activity levels of specific nuclides that cannot be readily measured by this method are often inferred using a radionuclide "fingerprint" [see below].

Radiochemical analysis of a representative sample set:
Representative samples are collected from waste materials within the waste stream of interest. The selected location, number and analysis requirements of these samples must be carefully considered to ensure that conclusions drawn from sample results are valid for extrapolation to the bulk waste activity. Waste stream fingerprints can be generated in this way to provide identification and relative activities of radionuclides which may be present in operational process wastes or decommissioning wastes. Many radiochemical laboratories are able to carry out determinations for a wide variety of radionuclides – both those emitting measurable gamma fluxes and those with only beta or low energy/intensity gamma emissions – allowing more comprehensive nuclide identification and quantification than offered by gamma spectrometry.

Historical data to be found in plant flowsheets, logbooks and event reports:
Historical data can be useful in showing the probable presence or absence of contamination in defined plant areas [e.g. from Health Physics survey data] which may guide the radiochemical sampling plan. This data may also indicate the specific radionuclide(s) to be expected from these plant areas.

Gamma Based Non-Destructive Assay

Low Resolution Gamma Spectrometry (LRGS)

Low Resolution Gamma Spectrometry systems are often based on the use of inexpensive and rugged sodium iodide [NaI(Tl)] detectors configured to view the whole or part of each waste item. These systems are often used to measure waste from relatively well characterized waste streams containing readily measurable radionuclides such as Co-60 and Cs-137, from which total radionuclide inventories are inferred using known radionuclide correlations [or "fingerprints"]. LRGS based systems are routinely used to measure low level radioactive waste arisings from several UK commercial power reactors and fuel cycle facilities (including uranium enrichment, fuel fabrication and reprocessing). The purpose of these measurements is to characterize the main gamma emitting radionuclides in the waste as a means of defining the most cost effective disposal route, while providing the mechanism, whenever practicable, to re-categorize waste for unrestricted release.

Correction for the effects of waste matrix attenuation of the detected gamma emissions is usually based on the waste item weight. Given a defined waste volume and measured weight the mean density of the waste matrix can be easily established. Correction factors can then be implemented for the detected gamma radiation of interest at the estimated waste density. This approach often assumes a degree of homogeneity for both the waste matrix and radionuclides but is applicable for both low and high density wastes where this assumption is reasonable. Practical issues are also important, for example for applications involving an uncontrolled environment sodium iodide based systems must incorporate gain stabilization due to the potential instability of the detector/system gain with variations in ambient temperature at the assay location.

The BNFL Instruments DrumScan®:LRGS systems incorporate all of these features in three basic configurations with two utilizing turntables for drum and package measurements, and the third forming a '4π' assay chamber for large waste components resulting from the decommissioning of nuclear power plants.

Figure 1 BNFL Instruments DrumScanâ : LRGS system in 200liter drum monitoring configuration and incorporating a separate workstation which can be located remote from the measurement unit to reduce operator dose uptake.

High Resolution Gamma Spectrometry (HRGS)

High Resolution Gamma Spectrometry systems are designed to measure a wide variety of wastes including those for unrestricted release, low level radioactive wastes, plutonium contaminated wastes, uranium contaminated wastes and fission and activation product contaminated wastes. Systems are configured to measure packages from 1 liter steel cans to $1m^3$ boxes, although 200 liter drum measurement systems are the most common configuration. The significantly better energy resolution of these systems compared to LRGS based measurements has two immediate advantages:

Poorly characterized waste streams containing highly variable mixtures of radionuclides are readily measured due to an ability to resolve individual gamma photopeaks within complex gamma spectra. Interpretation of spectral data then allows simple identification and quantification of the nuclides present using readily available compilations of gamma emission nuclear data.

A significantly greater number of radionuclides can be included in the data libraries of HRGS systems, allowing effective measurement of foreseen and unforeseen nuclides in complex spectra.

Both of these advantages lead to assay systems that depend less on alternative sources of data (eg flowsheet data and waste stream fingerprints) to achieve accurate assays.

Corrections for the effects of inhomogeneity in the waste matrix are usually based on one of the following three methods:

WEIGHT: The measured weight of the container and an estimate of the container fill height are used to calculate energy dependent, matrix correction factors.

DIFFERENTIAL PHOTOPEAK ATTENUATION: Measured count rate ratios from pairs of gamma lines from isotopes such as Cs-134, Co-60 and Eu-152, together with their branching ratios are used to calculate energy dependant matrix correction factors. Clearly this method does not provide a means of correction for all single emission energy radionuclides, eg Cs-137.

GAMMA TRANSMISSION: Gamma emission from a known transmission source located diametrically opposite the detector penetrates the matrix and is used to calculate energy dependent, matrix correction factors.

A new fourth method called "TransWeight" has recently been developed for high density, inhomogeneous wastes by BNFL Instruments [1]. This method combines the transmission and weight correction methods to provide accurate correction for partially filled containers of dense or stratified waste. Again, this method is used to calculate energy dependent, matrix correction factors. BNFL Instruments "DrumScan®:HRGS" systems incorporate these matrix correction techniques with the capability to automatically select the optimum correction algorithms for any waste container measurement or part container measurement (segment).

The overall versatility of HRGS measurements is highlighted by the use of DrumScan®:HRGS to measure pieces of ILW as they are removed from the Windscale Advanced Gas Cooled Reactor (WAGR) during decommissioning operations in the UK. Equally, DrumScan®:HRGS systems have been successfully combined with passive neutron coincidence counters to provide both quantitative uranium analysis and plutonium isotopic analysis in a fully integrated TRU monitoring systems.

Figure 2 BNFL Instruments DrumScanâ : HRGS in 200liter drum monitoring configuration

Case Studies

Sellafield Beta/Gamma Plant

BNFL operates the principal disposal site for solid low level radioactive waste (LLW) at Drigg in the UK. Disposal of waste to Drigg is from a wide range of facilities including power stations, isotope manufacturing facilities, universities and the nearby Sellafield nuclear fuel reprocessing plant. BNFL also operates a high force compaction facility for LLW in its Waste Monitoring And Compaction plant (WAMAC) prior to disposal at Drigg. As part of this operation, BNFL has put in place a comprehensive system of waste controls, part of which is a program of verification monitoring of waste on receipt from consignors. This results in both the individual LLW consignors and WAMAC carrying out gamma assay based measurements to confirm that waste is within the Conditions For Acceptance (CFA) of the Drigg disposal facility.

During the Spring of 1999 an inter-comparison exercise was carried out between HRGS and LRGS based systems for measuring waste consigned from a beta/gamma plant at BNFL Sellafield. This waste was first measured at the individual bag level using a DrumScan®: LRGS system and then as compacted, boxed waste using the WAMAC HRGS gamma scanner. The DrumScan®: LRGS employed incorporates two 3"x3" NaI detectors viewing each bag while rotated slowly on a turntable. Inspection of recent waste data from this beta/gamma plant indicated that Cs-137 and Co-60 were appropriate radionuclides for measurement with other nuclides being inferred from the waste stream fingerprints for the plant. Use of this single measured nuclide/fingerprint method to establish total activity values was later enhanced to direct measurement of <u>multiple</u> nuclides when it became clear that DrumScan®: LRGS spectral data could also yield accurate measurement of these additional nuclides. This change therefore resulted in a corresponding reduction on the dependence of fingerprint data. Appropriate spectral regions were set up in the DrumScan®: LRGS to provide a fully independent measure of the listed nuclides in each of the packages of waste.

Despite the generally penetrating nature of Cs and Co gamma radiation in mixed waste bags a further level of matrix correction (based on bag weight) ensured accurate assay.

Following measurement by the DrumScan®: LRGS in the beta/gamma plant this consignment of packaged low level radioactive waste was transported to WAMAC where it was carefully loaded into 1m3 (40" x 40"x 40") boxes and low force compacted. The resulting boxes were monitored using the WAMAC segmented gamma scanner. This system incorporates a single 30% HpGe detector which 'viewed' each box in a series of 11 segments of 4" depth. Correction for gamma ray absorption in the matrix material was based on either the waste weight or the differential photopeak attenuation methods. Measurement time for each box was 70 minutes.

Table 1. Summary of Results

Radionuclide	WAMAC HRGS (MBq)	DrumScan®: LRGS (MBq)
Cs-137	25.0	30.9±4.5
Co-60	0.5	0.5±0.1
Ru-106	13.8	11.1±1.9
Mn-54	12.9	8.2±1.4
Cs-134	2.4	1.5±0.3

Early observation of the archived waste stream fingerprints for this Sellafield beta/gamma facility yielded the following as major *measurable* radionuclides: Cs-137, Co-60, Ru-106, Mn-54 and Cs-134. The benchmark 'total consignment' activity for each of these nuclides was taken to be that provided by the WAMAC segmented gamma scanner. This was assumed due to the intrinsically enhanced ability of this system to identify/quantify radionuclide photopeaks together with confirmation of assay accuracy by extensive calibration checks [over the operating lifetime of WAMAC] and 'Round Robin' inter-comparison exercises.

The results [given above] were compared following DrumScan®: LRGS measurement of this entire consignment of packages and WAMAC HRGS measurement of the resulting boxes containing these packages. It can be seen that the consignment activities for all nuclides agree within approximately +/- 30% of the WAMAC values.

Clearly, direct measurement of these radionuclides by low resolution gamma spectrometry for as many measurable nuclides as possible can reduce the dependence of low level radioactive waste characterization on the availability & accuracy of waste stream fingerprints. However, the activity estimates of pure beta emitters such as H-3, C-14 and Sr-90 and radionuclides having low gamma energy/branching ratios will necessarily continue to depend on waste stream fingerprint data.

British Energy LLW Assay Systems

Routine operations in UK nuclear power stations give rise to low level radioactive waste that is consigned to BNFL's Drigg radwaste disposal facility. Whilst the volumes of waste consigned to Drigg from British Energy are relatively small, in order to reduce the volumes of waste still further, British Energy have introduced DrumScan®:LRGS systems to a number of their commercial nuclear reactor sites in the UK to support waste segregation and provide the necessary consignment data for waste disposal. The use of DrumScan®:LRGS systems was seen as a more cost effective [yet accurate] alternative to their traditional monitoring methods which, in many circumstances, were based on segmented gamma scanners. The features of these systems found to be particularly important by the station operators include:

An ability to assay drums, packages and HEPA filters with the same system.

Assay times of between 2 and 5 minutes to reach MDA's equivalent to the UK 'free release' limits

An ability to pre-select/optimize trial consignments by "mixing and matching" waste results from measured waste containers. Each pre-selected consignment predicts the consignment totals in terms of the waste disposal 'conditions for acceptance' and other consignment limitations.

Automatic production of the key consignment documentation with all radionuclide totals, de-minimis levels and consignment totals correctly calculated and inserted onto the shipping manifests for selected consignments.

Summary

Gamma based Non-Destructive Analysis techniques are well established as part of the overall management strategy for radioactive wastes arising from nuclear reactor operations and decommissioning. The factors relating to the choice of the most appropriate monitoring technique have been reviewed and ultimately depend on agreement being reached between waste consignor, regulatory authorities and waste receivers. In particular, decisions need to be made between the application of Low or High Resolution Gamma Spectrometries (LRGS and HRGS respectively) and the degree of reliance that can be placed on sampling and historical data in the assay process. Recognizing the diversity of specific requirements inherent in the application of such techniques to real applications, BNFL Instruments offer a comprehensive range of modular products utilizing both LRGS and HRGS techniques, such that the approach used can be readily tailored to meet individual customer needs. Within this family of products, the use of LRGS has emerged as an attractive alternative to HRGS for those applications in which the waste stream's radionuclide mix and matrix composition is relatively well characterized and is well suited to providing a cost effective route to utility waste management.

Refs:

'Improvements to Segmented Gamma Scanner Matrix Correction Techniques'; by P A Clark, D Parvin, K A Hughes, N Fitzgerald, J C B Simpson. INMM 40th Annual Meeting, 1999.

'An Inter-comparison of HRGS and LRGS based measurements of Sellafield Low Level Waste' ; by J Hoggarth and C H Orr. Cross Industry Working Group for Radwaste; July 1999