Mobile Assay System for Laboratory Standard Characterisation of Waste Drums and Larger Waste Objects – 25552

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ABSTRACT

In addition to radioactive waste drums there is a requirement to characterise larger objects such as waste boxes, waste pallets and waste bags, typically with a volume envelope in excess of a cubic metre. The measurement process is optimised if the system incorporates a heavy-duty turntable with a capacity typically in excess of 2 tonnes (2,000 kg), and also incorporating load cells for weight determination. Due to the size of the measured objects, such systems employ the Open Detector (OD) measurement geometry, where the detector views the entire sample object, and segmentation of the object is not employed. Generally, matrix density corrections for such larger object systems are performed by measuring the weight of the object, hence the load cells, and calculating an average density and applying a density correction on that basis. Inherent assumptions of such measurement systems are that the waste is homogeneous and the distribution of both density and activity is uniform throughout the waste matrix. The CHARMS (**Ch**aracterisation and **A**ssay **R**adioactivity **M**onitoring **S**tation) described in this paper is an example of an OD (far field) mobile assay and measuring system for making laboratory quality measurements of drums, waste bags and other waste objects in the field. It consists of a trailer with a laboratory/office and a heavy-duty turntable so that waste objects can be rotated to reduce the effects of inhomogeneity during a measurement. The detection system employs a high-resolution, high purity germanium (HPGe) detector with a large crystal diameter to enhance gamma ray detection. The HPGe detector is collimated and shielded to reduce the effect of the radiation background and radioactivity from other objects. The shielded detector system is mounted within the laboratory/office with a direct view of objects placed on the turntable for measurement. The motorised turntable, with an integral load cell for weight measurement, has a capacity of greater than 2 metric tonnes. The high efficiency (>50%) shielded and collimated HPGe detector with a large diameter crystal provides high resolution spectroscopic gamma ray measurements for radionuclide identification and activity determination. Minimum Detectable Activity (MDA) limits have been determined based on field measurements with a realistic background dose-rate. Using a 5-minute measurement time, MDAs of 0.0035 Bq/g for Cs-137 and 0.0021 Bq/g for Co-60 have been established. For measurements in lower backgrounds of Cs-137 and Co-60, and most other radionuclides of interest, CHARMS is able to sentence the waste into the various categories of Out-of-Scope, Very Low Level Waste (VLLW), Low Activity LLW (LALLW) and Low Level Waste (LLW). In order to qualify as an official laboratory measurement system under the ISO/IEC 17025:2017 measurement quality assurance standard, CHARMS has been subject to an intense assessment process conducted by the United Kingdom Accreditation Service (UKAS), the national government accreditation assessment body. "ISO/IEC 17025:2017 is the international standard for testing and calibration laboratories. It sets out requirements for the competence, impartiality, and consistent operation of laboratories, ensuring the accuracy and reliability of their testing and calibration results." ISO 17025 is widely applied in radiochemistry laboratories and portable radiometric instrument calibration facilities but is not widely adopted for waste package measurements on nuclear sites. The obvious benefit from adopting the requirements of ISO17025 is that the results obtained are robust and defensible. This paper describes the challenges of delivering laboratory-standard measurements on customer sites.

INTRODUCTION

A critical aspect of the growth in public support for nuclear power is public confidence that legacy nuclear facilities can be safely decommissioned and that the resulting radioactive waste can be stored and eventually dispositioned safely. Radioactive waste must not be seen as posing a risk to public health. The nuclear industry is almost unique in the efforts taken to control and manage radioactive waste, although this is not always understood by the public.

Improvements in radioactive waste measurement go some way to improving public confidence by quantifying, more accurately, legacy waste and by reclassifying waste from ILW and transuranic waste (TRU) to LLW and below. Reducing the amount of overclassified waste (TRU that is really LLW) not only saves cost but also reduces the perceived hazard associated in the public's mind with nuclear power.

In addition to improving radioactive waste measurement technology, a further positive step is to increase the demonstrated quality and provenance of radioactive waste measurement technologies. This can be achieved by requiring independently audited compliance with standards such as ISO/IEC 17025:2017, the measurement process standard currently a mandatory requirement for radioactive sample measurement laboratories in the UK. Currently, in both the US and the UK, only more general quality standards and local test standards are required for radioactive waste measurement. It has been apparent for some time that poor quality radioactive waste measurements, that have been controlled and documented to a high level of quality assurance, remain poor quality measurements. This is in part an explanation as to why there is so much overclassified waste in the US and the UK today.

Radioactive waste is stored, transported and sometimes measured in a variety containers ranging from cans, through drums, bags, boxes and large containers. Often overlooked is the truism that the effectiveness of a waste measurement is inversely proportional to the size of the container in which it is housed. This is particularly true for gamma ray measurements of radioactive waste, which are the most common.

It turns out that the 55 US gallon (200 litre) drum is a particularly effective container size for both neutron and especially gamma ray measurements of radioactive waste. A container with larger linear dimensions, for any reasonable waste matrix density, will have too much gamma ray attenuation. The effectiveness and sensitivity of radioactive waste measurements in drums have improved dramatically in recent years. The ANTECH WR-SGS (Wide Range Segmented Gamma Scanner) and the UDASS (Universal Drum Assay and Segregation System) are an example of this technology improvement [1,2,3,4,5,6]. These are both classified as Collimated Detector (CD) geometry instruments, and both incorporate transmission sources for matrix density correction and scanning to identify the distribution of radioactive sources in the waste matrix.

There remains a substantial problem measuring waste in larger containers, especially with the need for accuracy and the avoidance of waste overclassification. An initial issue is that virtually all measurements of radioactive waste in larger containers are performed using OD geometry or far field measurement technology. Unlike collimated detector instruments such as the WR-SGS or UDASS, for OD instruments the detector field of view includes the whole object being measured, be it a drum, bag or box.

Generally, the measurement involves obtaining a single emission spectrum for the waste object being measured. An advantage is that better counting statistics can be achieved compared to collimated detector instruments. The great disadvantage, however, is that no information is obtained on the distribution of density (and hence gamma ray attenuation) and the distribution of radioactive sources in the waste matrix. OD instruments are forced to assume that both the distribution of density and the distribution of sources is uniform and homogeneous. If this assumption is valid, then OD measurements can be very accurate. If the waste matrix is sufficiently heterogenous, in either density or source distribution, then large measurement errors will arise (7).

Associated techniques can be used to incorporate average density corrections when OD measurements are performed. The most common approach which is employed with CHARMS is to record the weight of the waste object (drum, bag or box) and calculate a gamma ray attenuation correction based on the average weight. A further approach, sometimes employed with SNAP (an OD waste measurement and analysis code) is to use differential attenuation, where a radionuclide emits gamma ray photopeaks at different energies, for example Co-60 and Pu-239. The average attenuation can be inferred from the differential attenuation of photopeaks arising from the same radionuclide. This approach is not as comprehensive as the use of a transmission source with a segmented measurement.

This paper describes the CHARMS OD geometry mobile laboratory standard measurement technology designed to improve the accuracy of waste object measurements in the field. It describes the process of testing to achieve accreditation to the ISO/IEC 17025:2017 measurement quality assurance standard and discusses measurement error analysis and the dangers of large errors arising in measuring large heterogeneous radioactive waste objects.

CHARMS MEASUREMENT SYSTEM

CHARMS is a mobile assay and radioactive waste measuring system for making laboratory quality measurements of drums, waste bags and other waste objects in the field. It is designated the ANTECH model G3650. Within the limitations and assumptions of OD measurement technology, CHARMS is designed to make accurate in-field measurements of a wide range of radionuclides in waste containers ranging from NORM (Naturally Occurring Radioactive Material) to fissile radionuclides in waste including both low and high activity. CHARMS is shown in use in the field in Figure 1 and 2 measuring different waste objects.

Fig. 1. CHARMS measuring a one cubic metre ‘builders’ bag’ in the field.

The typical measurement time for a CHARMS measurement is 5 minutes. Longer measurements may be selected to improve the MDA. The MDA depends on the local radioactive background as well as the density (attenuation) of the waste sample matrix and the radionuclides of interest in the measurement. Waste objects to be measured are normally loaded using a forklift.

The CHARMS measurement is normally performed using ORTEC Isotopic (Iso-Plus) measurement software, using the ORTEC IDM-200-V HPGe high efficiency detector, both of which are part of the accredited instrument. Although not included in the accreditation, other OD measurement software codes can also be used, such as the SNAP measurement software, or the ANTECH IsoCorr measurement software which incorporates density and geometry corrections.

Fig. 2. CHARMS measuring a 55-gallon (200 litre) drum.

CHARMS Mechanical and Electrical Components

CHARMS consists of a trailer with a laboratory/office and a heavy-duty turntable. The motorised turntable, with an integral load cell for weight measurement, has a capacity of 2 metric tonnes. CHARMS is designed to operate on 110 VAC, which may be supplied from 110 VAC mains or 240 VAC mains (through an isolation step-down transformer) or by on-board portable generator. Battery backup is provided as the CHARMS trailer is equipped with a 70 Wh backup battery (UPS) sufficient to keep the HPGe detector cooled down to operating temperature during non-operational times (power-cuts, nights, and weekends). A safety circuit is provided incorporating emergency stop buttons to stop rotation of the turntable.

CHARMS Detector

The CHARMS detection system employs a high-resolution, high purity HPGe electro-mechanically cooled detector with a large crystal diameter to enhance gamma ray detection. The HPGe detector, based on the ORTEC IDM-200-V, is collimated and shielded to reduce the effect of the radiation background and radioactivity from other objects. The shielded detector system is mounted within the laboratory/office with a direct view of objects placed on the turntable for measurement. The detector collimation does not restrict the detector field of view of the waste object so that the system sill employs OD geometry.

The high efficiency (>50%) shielded and collimated HPGe detector with a large diameter crystal (85mm) provides spectroscopic measurements with both excellent gamma ray energy resolution and sensitivity. Detection limits (MDA) have been determined based on field measurements with a realistic background dose-rate. Using a 5-minute measurement time, MDAs of 0.0035 Bq/g for Cs-137 and 0.0021 Bq/g for Co-60 have been determined. For measurements of Cs-137 and Co-60, and most other radionuclides of interest, CHARMS is able to sentence the waste into the various categories of Out-of-Scope, Very Low Level Waste (VLLW), Low Activity LLW (LALLW) and LLW.

CHARMS employs the OD or far field measurement method, as the entire waste object (drum, waste bag) is in the field of view of the detector. Using the weight measured by the load cells in the turntable (rotation platform) an average density attenuation correction is performed. During each measurement the waste object (for example drum or waste bag) is rotated at a constant rotation speed to reduce the effect if inhomogeneity in the waste matrix.

IMAGE Instrument

IMAGE is a separate instrument derived from CHARMS which is included in the CHARMS Accreditation. It consists of the CHARMS detector, the ORTEC IDM-200-V mounted on the scissor lift table dismounted from the CHARMS trailer, or more usually, mounted on an ANTCart. That configuration of IMAGE is shown in Figure 3. The measurement-control software is housed on the notebook computer also mounted on the ANTCart. Also shown in the figure is the Portable Drum Turntable loaded with a 55-gallon (200-litre) waste drum.

The ANTECH ANTCart is a rugged, portable detector trolley capable of holding a variety of detector types and lead shielding, including the Ortec IDM-200-V. The ANTCart features a detector platform that can be rotated, allowing the detector view to be pivoted through 180 degrees. It is positioned facing the side of the ANTCart to allow for ease of manoeuvrability and in order to allow the detector to be positioned as closely as possible to the container, if required. The detector platform can be raised and lowered using a hand driven linear actuator. The detector can be adjusted from a minimum height of 0.5m to a maximum height of 1.9m.

The portable drum turntable provides a rotational mounting platform for 200 litre (55 US galllon) drums and 340 litre (85 US gallon) overpack waste drums. The drum turntable is mounted on three high precision load cells, each rated at 300kg. Rotation is provided by a motor/gearbox slung under the portable drum turntable frame, connected to a control cabinet on the back of the assembly. The rotation speed is controlled by the motor inverter within the cabinet and can be adjusted using a USB connection to the operator laptop. Figure 3 also shows the portable drum turntable loaded with a 55-gallon (200-litre) waste drum.

As with CHARMS, IMAGE operates with ORTEC Isotopic (Iso-Plus) software, which is included in the accreditation. It can also operate with ANTECH IsoCorr density and geometry correction software.

Fig. 3. IMAGE may include the CHARMS detector (the IDM-200-V) deployed on the ANTCart mobile trolley.

LABORATORY ACCREDITATION

The use of general quality assurance standards, such as ISO9001 and NQA-1, as well as local measurement quality requirements, have failed to provide an adequate framework to ensure the integrity and quality of radioactive waste measurements, often leading to unquantified errors in measured radioactivity. In particular, they fail to provide either guidance or mandated requirements that form the link between the quality of measurement results and the measurement technology and measurement process required to deliver those results. Quality measurement results –'correct and reliable results'- are those with greater accuracy and reduced measurement errors. A further failure of current practice is that measurement physics constraints and critical assumptions about the measurement process often do not contribute to the measurement error assessment leading to potentially large and unquantified errors.

It has been apparent for some time that inappropriate and poor-quality radioactive waste measurements, despite being controlled and documented to a high level of quality assurance, still generate poor quality, incorrect results with large unquantified errors. This is particularly the case with OD instruments and the assumptions associated with measuring radioactive waste objects with unknown heterogenous matrices. It is the main reason that a significant quantity of TRU in the US and ILW PCM (plutonium contaminated material) and LLW in the UK has been overclassified.

A potential solution to this problem may be achieved by requiring independently audited compliance with measurement specific quality standards, such as ISO/IEC 17025:2017 [8], the international measurement and test standard. Currently, accreditation to ISO/IEC 17025:2017 is a mandatory requirement for radiochemical sample measurement laboratories in the UK. Arguable, it should be a mandatory standard for the measurement, quantification and sentencing of radioactive waste.

With these considerations in mind ANTECH chose to obtain ISO/IEC 17025:2017 accreditation in the UK for mobile laboratory measurement of radioactive waste drums and waste bags using CHARMS. In this way it is possible to demonstrate laboratory measurement quality to potential customers and differentiate CHARMS measurements from other OD measurements of radioactive waste objects.

The ISO/IEC 17025:1917 Standard

Quoting from the standard, "ISO/IEC 17025:2017 is the international standard for testing and calibration laboratories. It sets out requirements for the competence, impartiality, and consistent operation of laboratories, ensuring the accuracy and reliability of their testing and calibration results." ISO 17025 is widely applied in radiochemistry laboratories and portable radiometric instrument calibration facilities but is not widely adopted for radioactive waste package measurements on nuclear sites or in general in the nuclear decommissioning and radioactive waste cleanup industry.

The ISO 17025 requirements for measurement procedures ensure that the methods deliver the correct measurement results as they are validated techniques which are performed accurately by qualified staff whose training records are vetted. Measurement procedures are developed, verified and validated and subjected to expert technical scrutiny during the accreditation process and periodically thereafter. The scope of the measurement process is tightly defined and is directly related to the operating envelope which has been evaluated.

In the case of the CHARMS system, the scope includes the measurement of homogeneous radioactive waste samples in a variety of flexible intermediate bulk containers (FIBCs) (builders' bags) typically $1m^3$ and a variety of waste drums, typically 55 US gallon (200 litre). The scope includes a specific waste density range of 0.0012 - 1.65 kgm^{-3} and also includes gamma species within the energy range of 59 -1408 keV. Measurements which fall outside this envelope are not accredited. The measurement procedure starts by checking that the sample falls within the accredited parameters, and proceeds step by step to a tightly defined measurement report, with each step documented and (where they arise) non-conformances highlighted.

ISO/IEC 17025:2017 Testing and Validation

An initial requirement of the accreditation process is to perform blind measurement tests which produce satisfactory results. In the UK this includes a requirement to participate in the Nuclear Industry Proficiency Test Exercises (PTE) for drum measurements sponsored by the National Physical Laboratory (NPL). These involve a periodic scheme during which a 200 L drum of simulated waste is produced by NPL. This waste drum contains inactive low-density material in initially undisclosed locations spiked with gamma-emitting radionuclides of known activity. The drum is sent to participating labs, who then perform a 'blind' measurement of the drum using a non-destructive method.

The results of CHARMS recent participation in a blind test drum measurement as part of the PTE laboratory intercomparison scheme are displayed in Figure 4. CHARMS measured the specific activity for the three included radionuclides and the results are plotted against the NPL declared activity values, which are supplied after the measurements are submitted. Even for a low-density matrix the 59 keV gamma rays from Am-241 are more difficult to measure, reflected in the larger error bars.

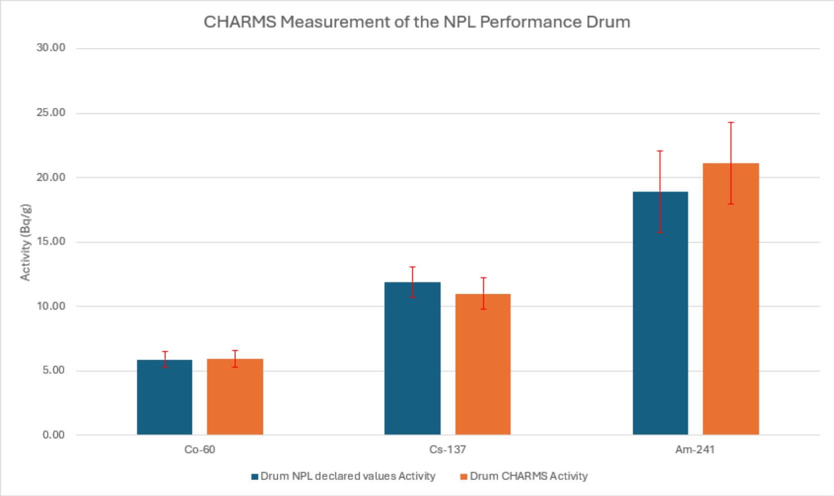

Fig. 4. Result data for the measured activity from a blind test drum measured a part of the PTE laboratory intercomparison.

For gamma ray instrument testing and measurement process validation, ANTECH have developed a series of test drums for measurement systems. These are based on the use of Europium-152 line or rod sources placed within re-entrant tubes into test drums, positioned in a helical array such that when rotated, each line source describes an annulus of activity, spaced such that each annulus represents an equal volume of the hypothetical true homogenous volume source. When viewed by a gamma ray detector, the drums appear to have a radioactive matrix which is homogeneous.

The line sources are surrounded by a homogeneous inert attenuating matrix, with a choice of matrices in different drums covering a density range of 0 - 1.65gcm^{-3}; using air, sawdust, water, and sand. This accurately simulates actual waste containers and is thus used variously as a step in calibration, verification or validation of ANTECH gamma ray-based drum radioactivity measuring systems. A typical simulated waste drum with re-entrant tubes with rod sources is shown in Figure 5. In the photo the rod sources are inserted into the tubes and one of the rod sources is visible lying on the top of the drum.

Fig. 5. This photograph shows a test drum with re-entrant tubes into which rod sources can be placed. Also shown is a view of a test bag on the CHARMS turntable. Note the location of re-entrant tubes inserted into the bag, into which rod sources may also be placed for testing.

When seeking accreditation for measurements of FIBCs (builders' bags), a similar test configuration was required. Figure 5 shows a simulated waste bag on the CHARMS turntable with a wooden frame structure supporting re-entrant tubes into which sources can be inserted for testing. The positioning of the re-entrant tubes goes some way to simulating a uniform matrix when the bag is rotated, although not as well as can be achieved by the more regular cylindrical drum geometry.

Figure 6 displays the results of CHARMS measurements of simulated waste bags – FIBCs populated with different fill heights of Eu-152 rod sources. The sample (declared) activity and the average measured activity are compared.

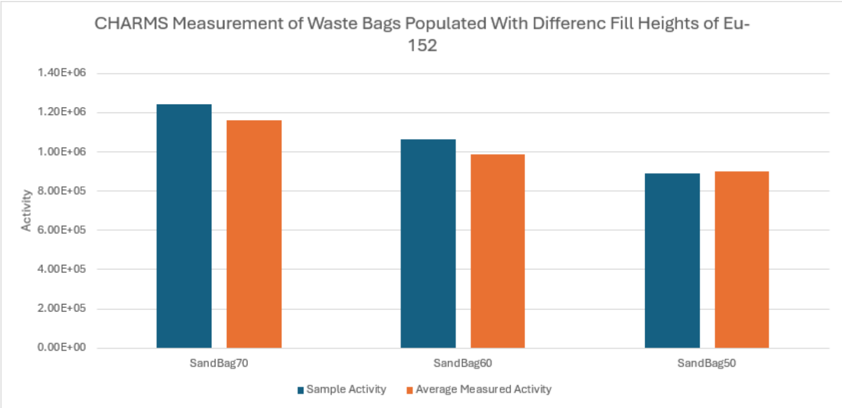

Fig. 6. CHARMS measurements of simulated waste bags populated with different fill heights of Eu-152 rod sources

The availability of measurement data from these systems means that ANTECH can produce an internal intercomparison of systems that have measured these test drums. This intercomparison contributes to the wealth of evidence that together validates and verifies the measurement instructions AIMS-MI-8602 and AIMS-MI-8603 for measurement of waste containers using the CHARMS system. AIMS documents are formally part of the ANTECH Integrated Management System, which encompasses quality assurance activities.

During the accreditation assessments and re-certification audits, evidence for compliance with the ISO 17025 requirements is scrutinised by trained assessors and subject matter experts. The assessments include witnessing live performance of measurements to ensure that the measurement instructions are being followed correctly.

ANTECH has found that the ISO 17025 standard gives a comprehensive framework for the development and operation of a high-quality measurement service and accrediting measurement services to the standard gives customer added confidence. The standard defines the term of "laboratory" not as a place, but as the body that performs the testing activities, and as such, it is easily adaptable for site working on nuclear licensed sites.

DEMONSTRATION MEASUREMENT

A trial at the UK's Low Level Waste Repository (LLWR) was held under the Nuclear Waste Service (NWS) Integrated Waste Management Programme (IWMP). An objective of the IWMP is to deliver improved characterisation techniques required to support the Nuclear Decommissioning Authority (NDA) Group's decommissioning mission. NSG Environmental Ltd (NSG), as a waste characterisation services framework holder, were contracted to assess the suitability and pilot the use of novel assay and characterisation systems for on-site radiological assay / characterisation of bulk soil/ excavation wastes. The ANTECH CHARMS system was assessed under this contract. The measurements took place at the LLWR site near Drigg in Cumbria in the UK.

The waste soil was in FIBCs, (builders' bags) and was measured in a short trial. Twenty bags of soil and excavation waste were measured with an average activity of 1.72Bq/g (standard deviation 0.33Bq/g). Almost all of the activity in the bags was attributable to naturally occurring radioisotopes, with potassium 40 contributing 73% of the activity as well as contributions from the U-238 and Th-232 decay chains.

Measurements were carried out with 600s and 900s measurement times, and the MDAs calculated for each isotope. The results are displayed in Table I.

Table I, MDAs calculated for soil and excavation waste at the LLWR

		900s MDA	600s MDA
Nuclide	**Energy (keV)**	**Bq/Kg**	**Bq/Kg**
K-40	1460.54	20	30
Ac-228	337.43	74	86
Cs-137	660.64	5.3	6.6
Bi-214	1764.68	25	31
Tl-208	510.89	37	40
Pb-214	295.39	58	69
Am-241	58.8	111	152

After the rebuilding and re-commissioning of the CHARMS instrument, the trial was a useful test of the system, especially the practical aspects of mobilisation to and demobilisation from a remote site. The trial has provided an opportunity to review the measurement process (in keeping with ISO/IEC 17025) and in the light of different customer and site reporting requirements. The revised results data report will include at a minimum:

1. Sample or package ID
2. Gross sample mass (kg)
3. Net sample mass (kg)
4. Total sample activity (MBq)
5. Total sample activity per unit gross mass of non-alpha emitting radionuclides (MBq/kg)
6. Measured sample gamma ray spectra
7. Measurement time
8. Calculated bulk density (kg/litre)
9. Calibration file
10. Library of isotopes employed
11. Calculated MDAs for the library (including measurement set-up parameters used in the calculation)
12. Tracking Log (containing actual measurement times and sample or bag weights)
13. Specific activity file (contains specific activity for each of the isotopes reported in the library)

CONCLUSIONS

This paper has described CHARMS, an open detector based mobile assay and radioactive waste measuring system for making laboratory quality measurements of waste drums, waste bags, waste boxes and other waste objects in the field. It is designed to have enhanced measurement performance by including a high efficiency well shielded HPGe detector on an efficient mobile platform with a heavy-duty turntable and built in loadcells for sample weight measurement.

Measurement results from drum measurement performance tests of the NPL PTE drum are reported for radionuclide measurements of Co-60, Cs-137 and Am-241. Measurements of simulated FIBCs, (builders' bags) with an approximately uniform source distribution of Eu-152 sources of different bag fill heights are also reported. Good agreement is achieved comparing the declared and measured

activity. Calculated MDA values are also reported from measurements of typical soil and excavation waste in FIBCs for several radionuclides of interest.
The case for performing ISO/IEC 17025 accredited measurements is presented as a means of improving the quality of radioactive waste measurements and avoiding waste overclassification. The dangers of Open Detector measurement instrument misuse are emphasized as is the consequent risk of large and indeterminant measurement errors.

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