Reclassification of Intermediate Level Waste (ILW) Using the ANTECH UDASS – 23493

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ABSTRACT

Evidence has been available for some time both in the UK and the USA that a significant fraction of Intermediate Level Waste (ILW) in the UK and Transuranic Waste (TRU) in the US has been misclassified over a period of many years. In part this is due to the conservative assumptions associated with measurements of ILW. It is also due to the often-inappropriate use of Open Detector based assay systems that require the assumptions that the radioactive waste is homogeneous in both density and activity distribution. This leads to large measurement errors and an over estimation of waste activity. A similar outcome arises when the radiological classification is based on Nuclear Safety Value measurements, again based on conservative assumptions. In order to overcome this difficulty, a drum-based assay system has been developed incorporating increased sensitivity and the improved ability to correct for inhomogeneity in the distribution of both matrix density and radioactivity within a waste drum. The new system has been designated the Universal Drum Assay and Segregation System (UDASS) and is designed with the purpose of assigning waste drums to the lowest relevant radiological category by minimising measurement uncertainties. The UDASS is based on the use of a single high efficiency high purity Germanium (HPGe) detector that can be operated in either Open or Collimated detector mode. The technology has evolved from earlier instruments and encompasses an advanced variant of the Open Detector method known as the Dynamic Open Detector (DOD) method, the Wide Range Segmented Gamma Scanner (WR-SGS) method and finally the Tomographic Gamma Scanner (TGS) method employed in transmission mode. All three assay techniques or methods are implemented in the single UDASS platform. In order to determine the optimum approach to the assay of a given waste drum, emission and transmission Pre-Scans are performed using the WR-SGS method. If a drum is sufficiently homogeneous then a DOD emission measurement is performed. If the drum is assessed to be heterogeneous in either density or activity distribution, then a WR-SGS emission measurement is performed. The results of the pre-scan transmission measurement are used to perform a transmission correction to adjust the results for matrix attenuation for either DOD or WR-SGS emission scans and produce a preliminary assay result. If a WR-SGS assay result falls close to a radiological classification boundary a TGS transmission measurement may be undertaken to improve the attenuation correction. Following extensive commissioning and Factory Acceptance Testing, which is described in the paper, the UDASS was deployed to the site of the UK Low Level Waste Repository (LLWR) operated by Nuclear Waste Services, a subsidiary of the UK Nuclear Decommissioning Authority (NDA). At LLWR, 1,810 ILW PCM (Plutonium Contaminated Material) drums from earlier site decommissioning activities were stored, with no clear disposition path. The UDASS was tasked with re-measuring the 1,810 drums as part of the site PCM Drum Re-characterisation Project. The project has been a remarkable success. More than 95% of the ILW drums have been reclassified as either LLW or below LLW. Of these a total of more than 85% have been reclassified as below LLW facilitating their disposal to appropriate UK licensed landfill sites thus diverting the waste away from LLWR where disposal space is at a premium. The assessed radiological hazard of the LLWR site has been significantly reduced and drum storage and disposal cost savings of more than nine million pounds have been achieved.

INTRODUCTION

In both the UK and the US, significant quantities of assumed higher activity plutonium bearing waste have been accumulating for many years, posing a very costly disposal problem in the future. And yet, it has been evident for some time that a significant fraction of this higher activity radioactive waste had been misclassified. The waste in question is the Intermediate Level Waste with plutonium contaminated material (ILW-PCM) in the UK and Transuranic Waste (TRU) in the US. The misclassification has been largely due to two factors.

The first is the use of conservative assumptions associated with measurement processes for ILW and TRU waste. The second is due to the often-inappropriate use of Open Detector based assay systems that require the assumptions that the radioactive waste is homogeneous in both density and activity distribution. The use of these assumptions often leads to large measurement errors and a consequent over estimation of waste activity. A similar outcome arises when the radiological classifications is based on Nuclear Safety Value measurements, again based on conservative assumptions.

In order to overcome this difficulty, a new waste assay system, designated the Universal Drum Assay and Segregation System (UDASS), has been designed with the purpose of assigning waste to the lowest relevant radiological category. To achieve this, the components and their configuration have been carefully selected to minimise systematic measurement uncertainties. The UDASS is a drum-based system and has been developed incorporating increased sensitivity and the improved ability to correct for inhomogeneity in the distribution of both matrix density and radioactivity within a waste drum.

The UDASS has had an evolutionary development and incorporates a variety of drum measurement methods. The simplest of these is the Open Detector method, where the entire drum is in the detector field of view. The Open Detector (far field) Geometry method [1] remains useful for initial screening of drums and to provide an assay result for those drums that are homogeneous. The situation for heterogeneous drum screening improved with the advent of the Segmented Gamma Scanner (SGS), which was developed by Jack Parker and Ray Martin [2] at Los Alamos in 1977. Using a transmission source, the SGS makes a separate density correction for each fixed vertical segment of a drum and represents a significant but limited improvement over measurements using open detector geometry. A further potential improvement arose in 1995 when Robert Estep and others at Los Alamos developed the Tomographic Gamma Scanner (TGS). The TGS measures drums in relatively small volume elements (voxels). Although useful for many waste assay applications and some transmission measurements, poor counting statistics render the TGS generally inapplicable to LLW – ILW (PCM) or LLW – TRU segregation.

In 2009, and with considerable experience in building SGS [3] and TGS [4] instruments, ANTECH devised the Wide Range SGS to extend the range of applicability of the SGS technique. The extension encompassed both high and low density and high and low activity drums. The three essential features that distinguish the WR-SGS from conventional SGS instruments are a variable aperture collimator (VAC), helical (continuous) scanning in place of discrete scanning of each segment and a transmission source that can be placed in a shielded safe when not required. The first two features overcome the limitation of fixed drum segments which opens the way for much more effective measurements for LLW – ILW (PCM) or LLW - TRU segregation. ANTECH has several WR-SGS systems [5, 6, 7, 8] operating around the world, all with different configurations for different applications but all with the basic common features. They are used to measure a range of low to high activity drums, which can vary from very low density to very high density.

In 2018 the WR-SGS design was extended with software and hardware enhancements to measure drum density profiles and drum activity profiles of suspected ILW (PCM) or TRU drums, using an emission and a transmission pre-scan procedure, as a first stage of the drum screening process. At the same time the platform was extended, first to incorporate the ability to perform enhanced Open Detector measurements for homogeneous drums, and second to facilitate TGS transmission measurements, providing voxel by voxel enhanced gamma-ray attenuation corrections as an option to improve the attenuation correction employed with the SGS emission measurements. TGS operation was enabled by the inclusion of a drum translation axis, although only TGS transmission is implemented due to the counting statistics problem with TGS emission measurements. This novel integration of proven measurement methods incorporating enhanced Open Detector Geometry, WR-SGS and TGS transmission measurement capability has been designated the ANTECH Universal Drum Assay and Segregation System (UDASS). A typical UDASS is shown in Figure 1 with a 200-litre drum in the measurement position.

Fig. 1. UDASS with 200 litre waste drum in the measurement position.

MEASUREMENT METHODS

The UDASS development has been controlled under the ANTECH certified ISO 9001:2015 Quality Assurance (QA) system, (certificate FM36310), which also encompasses the relevant requirements of NQA-1. The measurements and operation of the UDASS is in keeping with the requirements of the measurement QA standard ISO/IEC 17025:2017 for laboratory testing and calibration. ANTECH is currently the only UK organisation with UKAS Accreditation under ISO/IEC 17025:2017 (Registration No. 10301) for drum and waste bag measurements of radioactive waste using the ANTECH instruments, CHARMS and IMAGE. ANTECH has recently (February 2023) achieved UKAS Accreditation under ISO/IEC 17025:2017 for the UDASS. This process is equivalent to testing laboratory accreditation in the USA under the National Voluntary Laboratory Accreditation Program (NVLAP) and DOELAP.

The UDASS is based on the use of a single high efficiency high purity Germanium (HPGe) detector that can be operated in either Open or Collimated detector mode. It is used in combination with an Eu-152 transmission source which is employed for drum matrix density determination. The technology has evolved from earlier ANTECH instruments and encompasses an advanced variant of the Open Detector method known as the Dynamic Open Detector (DOD) method, the Wide Range Segmented Gamma Scanner (WR-SGS) method and finally the Tomographic Gamma Scanner (TGS) method employed in transmission mode. All three assay techniques or methods, including the Pre-Scans, are implemented in the single UDASS platform.

Although the UDASS is directed towards low activity measurements associated with ILW – LLW (or TRU – LLW) segregation, it has the capability to measure very high activity drums. In combination with the VAC, (which can be used to reduce the input count rate), the UDASS also incorporates an automated tungsten filter for measurements of high activity drums where the drum gamma ray dose rate might saturate the detector and counting system. A typical drum measurement cycle time for both DOD and WR-SGS measurement modes is 34 minutes, which includes drum loading, Pre-Scans and the assay measurement, and drum unloading. If a TGS transmission measurement is added, then additional measurement time is required of typically a minimum of 20 minutes.

Emission and Transmission Pre-Scans

Prior to a drum measurement, both emission and transmission Pre-Scans are performed using the WR-SGS method. These assess the degree of homogeneity of the waste drum matrix and in the distribution of radioactive sources in the waste. Based on the results of the Pre-Scans, the UDASS decides if the drum is sufficiently homogeneous to employ the DOD measurement method or, if the drum is heterogenous, to employ the WR-SGS measurement method. The results of the pre-scan transmission measurement are used to perform a transmission correction to correct the results for matrix attenuation for either DOD or WR-SGS emission scans and produce a preliminary assay result. Once automatically selected, either a DOD or a WR-SGS measurement is undertaken, and an initial assay result is calculated. WR-SGS is the default option. Plotted Pre-Scan results are available to the operator, who can override the automated assay method selection if appropriate.

Dynamic Open Detector

The DOD method is selected for those drums deemed to satisfy the criteria for matrix and source distribution homogeneity. For drums which meet this criterion, the DOD method delivers the lowest counting uncertainty and therefore the best possible measurement result. It represents a significant improvement over conventional Open Detector measurement technologies. It is based on helical scanning of the drum with a large VAC opening and the emission data acquisition process employs list mode. The DOD emission data is acquired in segments which are a subset of the segments used in the Pre-Scan transmission measurement. The segment-by-segment emission data is then attenuation corrected using a combined version of the Pre-Scan transmission data. Traditional Open Detector spectra are normally attenuation corrected, not on a segment-by-segment basis, but using a single attenuation (density) factor calculated from the drum weight and not considering density variations within the volume of the drum.

Open detector measurements performed using far field geometry tend to have small geometric errors arising from drum end effects. However, gamma ray detection efficiency decreases in a non-linear manner as the detector moves further from the drum. Traditional Open Detector measurements are often made in near field geometry in close proximity to a drum to optimise detection efficiency. This may lead to large and uncorrected end effect errors. In the DOD method, these potentially large end effect errors are corrected as a result of acquiring the emission data in segments.

Wide Range Segmented Gamma Scanner

The WR-SGS measurement method has extensively been described elsewhere [5, 6, 7, 8]. The method is employed in the Pre-Scan and in the emission scan if the WR-SGS assay method is selected when a waste drum is deemed heterogenous. The Transmission scan performed as part of the Pre-Scan is combined with the WR-SGS emission scan to provide the attenuation correction and the assay result from the WR-SGS method. As with the Pre-Scans and the DOD emission scan, all WR-SGS emission and transmission scans employ list mode data acquisition.

Tomographic Transmission Segmented Gamma Scanning

As with the WR-SGS, the ANTECH implementation of the TGS method has been described elsewhere, [3]. In the UDASS only TGS transmission has been implemented and the method is designated the TTSGS as it is employed in conjunction with a WR-SGS emission measurement. As well as improving the attenuation correction for highly heterogenous waste drums, the TTSGS method enables enhanced visualisation of drum density variations (solid objects). In TTSGS mode the system also performs in a manner similar to a low-resolution X-ray scanner for investigating drum matrix anomalies. The TTSGS attenuation correction improves as the TTSGS measurement time is extended. This is also true for the TTSGS images of drum density variation. In exceptional cases, perhaps as part of a specific drum investigation, long measurements of higher resolution (more segments resulting in smaller voxel dimensions) may be made resulting in very detailed attenuation maps for each segment of the drum. This approach could be useful when investigating an anomalous object in a drum.

TESTING and VALIDATION

The UDASS has been subjected to rigorous and comprehensive testing and verification. Following extensive computer modelling, setting to work tests were conducted by ANTECH, including measurement and calibrations employing standard test drums with different matrices into which 6 Eu-152 standard rod sources, arranged in a helical array, are inserted to simulate uniform source distributions in drums. Calibrations have been performed based on both MCNP computer modelling as well as measurements of radioactive sources traceable to international standards. A comprehensive and very flexible complex test drum was designed and constructed with modular wedge-shaped matrix components of differing densities into which rod or point sources may be inserted. The test drum was used for initial UDASS testing and is employed as part of the continuing testing and development of the UDASS technology.

In 2020 ANTECH was awarded a measurement service contract by the UK Low Level Waste Repository (LLWR), to measure 1,800 legacy ILW-PCM drums stored on the site with no obvious disposal path. LLWR is now a part of Nuclear Waste Services, a subsidiary of the Nuclear Decommissioning Authority (NDA) a UK government body responsible for decommissioning UK nuclear facilities and UK nuclear liabilities, including nuclear waste. The Sellafield site (SL) is also a subsidiary of the NDA.

Under the terms of the LLWR measurement service contract, a Sellafield based independent physics assessor was appointed to oversee, witness, and assess the Factory Acceptance Testing (FAT), subsequent on-site Customer Acceptance Testing and review the ongoing measurements of LLWR ILW-PCM radioactive waste drums. As part of this independent oversight and review process, ANTECH have undertaken a comprehensive calculation and estimation of UDASS systematic measurement errors. Error analysis modelling data was also used from an earlier ANTECH study [9]. UDASS minimum detectable activity (MDA) calculations based on the ISO-11929-3 standard were also reviewed and combined with the systematic error data to establish definitive measurement errors for UDASS measurements. The calculations, estimates and assessments have been reviewed by the SL based independent physics assessor supported by additional SL physics staff and approved. With this approval LLWR staff are able, based on UDASS measurement results, to consign the much of the ILW-PCM waste to lower radiological categories.

In addition to the physics and measurement quality assessments for the UDASS, LLWR have undertaken a comprehensive review of ANTECH software development and control procedures. These procedures have also been reviewed by the United Kingdom Accreditation Service (UKAS) as part of an assessment of the UDASS for accreditation under ISO/IEC 17025:2017 for laboratory testing and calibration. This independent third-party assessment has also involved compliance of measurement and calibration processes

to ISO/IEC 17025 as well as a thorough examination of a variety of aspects of UDASS operation, ranging from operator qualification and training to an assessment of measurement results from blind tests.
To illustrate both the testing and operation of the UDASS, the measurements of two standard test drums, are reported. The first test drum was homogeneous (of uniform density and activity), and the other drum was inhomogeneous (with a complex matrix structure and non-uniform radioactive source distribution).

Homogeneous Drum Test

In order to test the DOD method a homogeneous (uniform) test drum was constructed. Six standard rod sources were placed in re-entrant tubes arranged in a helical array in a sawdust filled drum to simulate a uniform drum matrix with a uniform activity distribution. Following the Pre-Scan measurements, the UDASS confirmed drum homogeneity and selected the DOD measurement and analysis method. Figures 2 and 3 display plots of the pre-Scan transmission and emission data based on the integrated count rate.

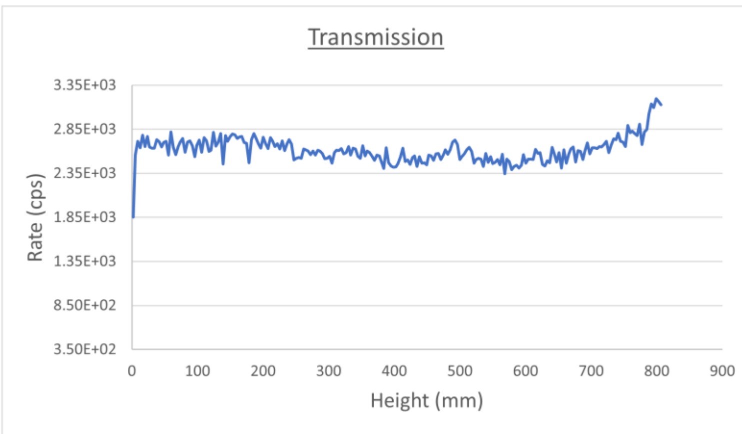

Fig. 2. Pre-Scan transmission integrated count rate data plotted as a function of vertical position in the homogeneous test drum. The lower transmission at the base of the drum (0 mm) is due to absorption in the metal drum base. Variations of attenuation are due to settling of the contents over time. Higher transmission at the top (800 mm) is due to a gap in the sawdust matrix material.

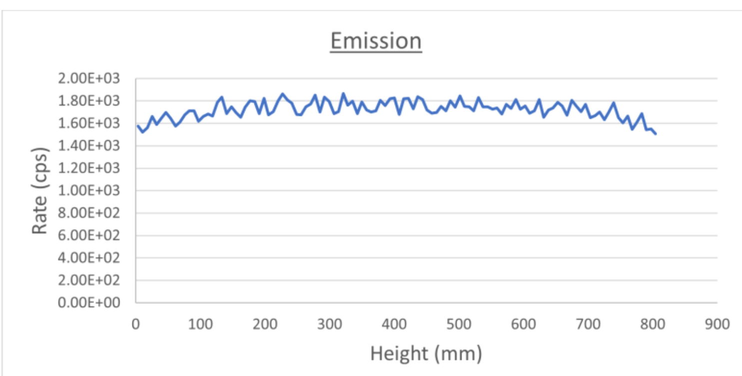

Fig. 3. Pre-Scan emission integrated count rate data plotted as a function of vertical position in the homogeneous test drum. The reduced count rate at either end is a result of the wide detector field of view being restricted at the top and bottom of the drum. The UDASS compensates for this effect.

The transmission Pre-Scan employed a 4mm collimator aperture and a 32-segment scan. The emission Pre-Scan employed a 64mm collimator aperture and an 8-segment scan. The drum passed both the emission and transmission test for homogeneity. The DOD analysis used the measured transmission information from the Pre-Scan to determine the average transmission for each of the 8-segment employed in the emission scan. The result of the DOD measured activity is tabulated in Table I.

Table I, DOD Measurement Results

Nuclide	Activity (Bq)	Total 2 sigma Uncertainty %	Ref. Activity (Bq)	Deviation from actual %
Eu-152	1.03E+06	35.3%	1.16E+06	-11%

Inhomogeneous Drum Tests

A much more complicated test drum was constructed in order to test the ability of the UDASS to correctly assay distributed sources in heterogenous matrices. Modular components of the flexible complex test drum were employed in constructing the drum for the UDASS inhomogeneous drum test. For the test the drum was divided into 4 sectors from top to bottom.

The top sector consisted of two high-density polyethylene objects (letters), the letter 'T' above the letter 'S'. This sector is to test the TTSGS ability to enable the visualisation of solid objects. The second sector consists of 2 layers of cork wedges which contain a single Am-241-point source. This sector is to test the UDASS ability to locate and quantify point sources in a matrix. The third sector consists of two complete layers of high-density polyethylene wedges with uniformly distributed Eu-152 rod sources. The third sector is to test the UDASS ability to identify and quantify distributed sources in a higher density matrix. The fourth and bottom section consists of two complete layers, half of which are high-density polyethylene wedge shaped sections and half of which are cork wedge shaped sections. The sector contains distributed Eu-152 rod sources. The bottom sector is to test the ability of the TTSGS method to provide a detailed matrix density map revealing the fine detail of density variations for the sector.

The Pre-Scan integrated count rate transmission and emission data is plotted in Figures 4 and 5 as a function of vertical position in the inhomogeneous test drum.

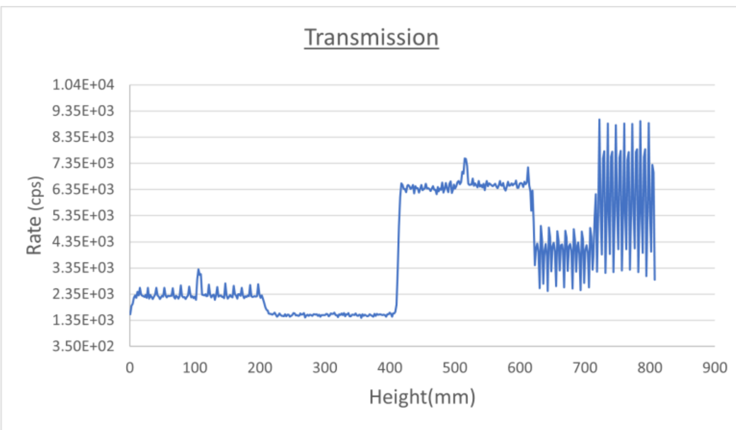

Fig. 4. Pre-Scan transmission integrated count rate data plotted as a function of vertical position in the inhomogeneous test drum. The division into 4 sectors is clearly visible. In the top sector, the count rate varies as the high-density polyethylene letters are rotated. Other transmission 'spikes' reveal the small gaps between wedge sections.

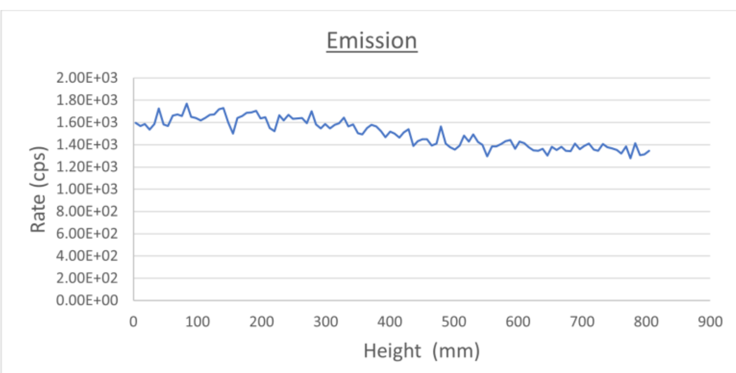

Fig. 5. Pre-Scan emission integrated count rate data plotted as a function of vertical position in the inhomogeneous test drum.

Figure 6 is a double plot of the measured activity per emission segment (8) and the transmission integrated count rate per transmission segment (32) for the inhomogeneous test drum.

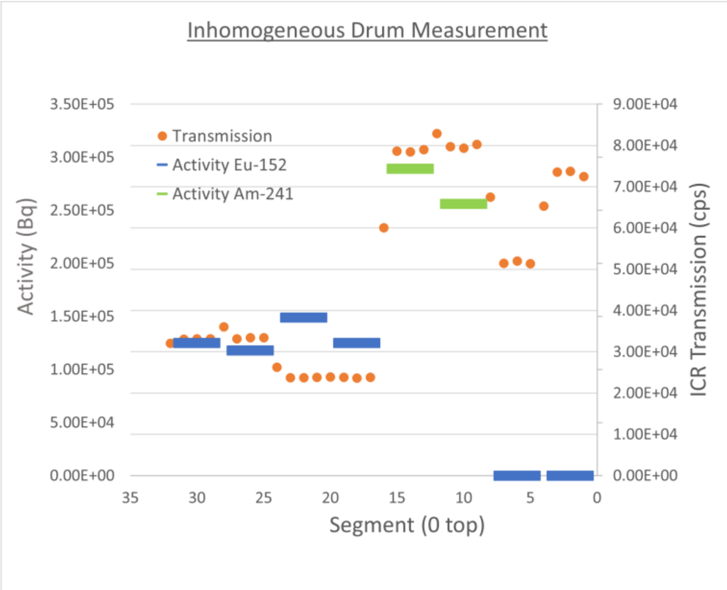

Fig. 6. Double plot of the measured activity per segment and the transmission integrated count rate per segment for the inhomogeneous test drum. The drum matrix structure is again apparent from the diagram. The activity (for Eu-152 and Am-241) for each of the eight segments in the WR-SGS emission scan is displayed.

The UDASS correctly identifies the distributed Eu-152 source and the Am-241-point source in the different regions of the drum. The Am-241 source is detected in two of the wide emission segments. A more precise location of the point source would have been made if a larger number of smaller emission segments had been used with a smaller collimator aperture. No sources were found in the top sector of the drum where the high-density polyethylene letters are located.

The density (attenuation) map produced by the optional TTSGS measurement is displayed in Figure 7, below. For clarity some features have been highlighted in red. At the top of the drum the letter 'T' and below it the letter 'S' can be seen. In sector 8 at the bottom of the drum the half drum high density sections (high-density polyethylene) are clearly distinguished from the half drum low density sections (cork).

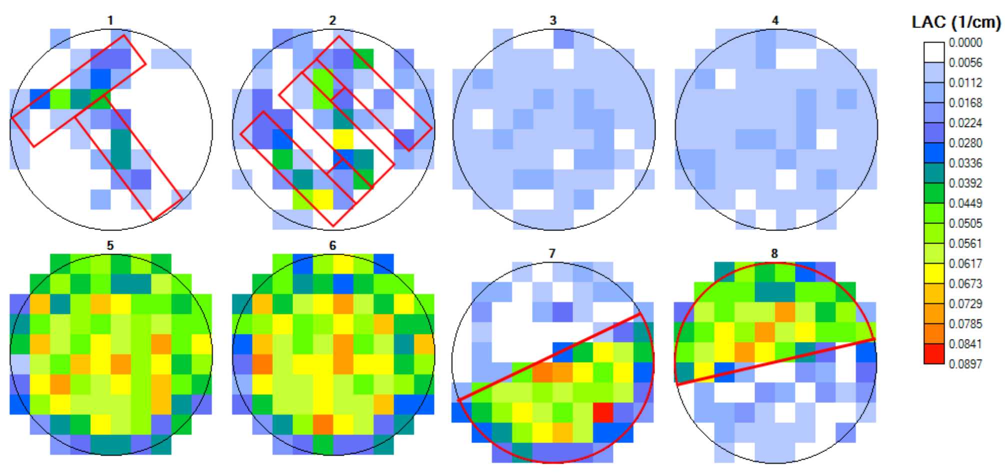

Fig. 7. TTSGS generated images (map) of the test drum density variations. Segment No. 1 is at the top.

For the complex inhomogeneous test drum, the transmission Pre-Scan employed a 16mm collimator aperture and a 32-segment scan. The emission Pre-Scan employed a 64mm collimator aperture and an 8-segment scan. The drum failed the transmission test for homogeneity, so a WR-SGS measurement was selected. The WR-SGS analysis used the measured transmission information from the Pre-Scan to determine the average transmission for each of the 8-segment employed in the emission scan. The results of the WR-SGS measurement are in agreement with the known characteristics of the drum and the measured activities are tabulated in Table II and compared to the reference activities.

Table II, WR-SGS Measurement Results

Nuclide	Activity (Bq)	Total 2 sigma Uncertainty %	Ref. Activity (Bq)	Deviation from actual %
Eu-152	5.38E+05	88.3%	5.81E+05	-7%
Am-241	5.24E+05	88.5%	4.27E+05	23%

MEASUREMENT OF ILW DRUMS AT LLWR

As part of the site PCM Drum Re-characterisation Project, a measurement service contract was awarded to ANTECH in 2021 by the UK Low Level Waste Repository (LLWR). This was for the UDASS to measure 1,810 legacy ILW-PCM drums stored on the site from earlier site decommissioning activities. The drums had been classified as ILW following previous Nuclear Safety Value measurements made using a less sensitive assay instrument employing conservative assumptions. The drums had no obvious disposition and disposal path except transport to the Sellafield Site for indefinite storage.

Following extensive commissioning and Factory Acceptance Testing, which is described in the paper, the UDASS was deployed to the LLWR site. The measurement campaign lasted a total of 15 months during which over 1,810 ILW drums were measured, and the results analysed and reported to LLWR. At the outset of the campaign, it was considered that as many 40% of the drums might be reclassified from ILW to LLW. In reality, the UDASS has achieved a reclassification rate of over 95%, as shown in Figure 8.

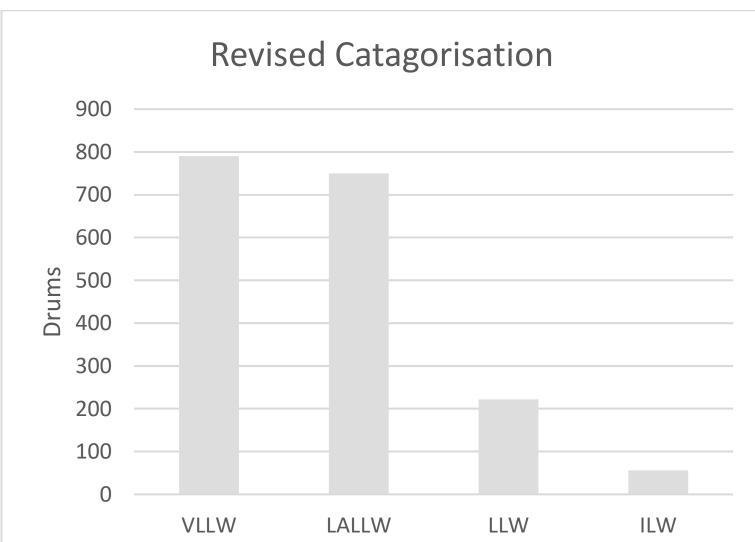

Fig. 8. The revised distribution of drum radiological categories following drum reclassification as a result of the UDASS measurements of ILW-PCM drums at LLWR. The number of drums is plotted as a function of radiological category.

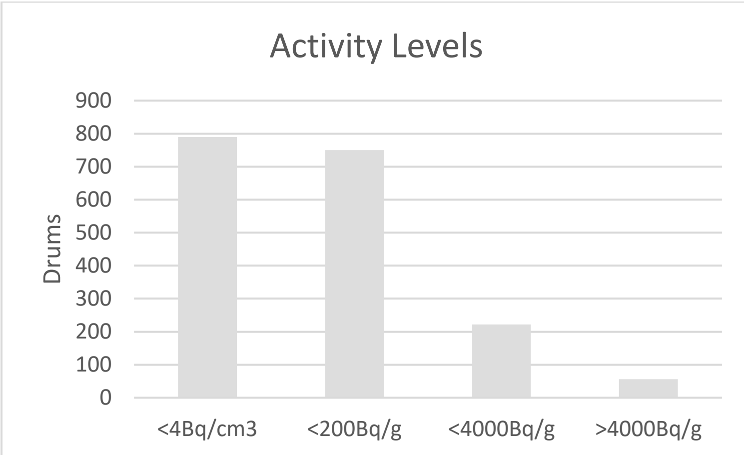

Fig. 9. The distribution of total activity for measured LLWR drums is displayed. The bar chart tabulates the number of drums with specific activity below a designated threshold.

Of the drums reclassified from ILW, a total of 85% have been reclassified to the UK radiological categories below LLW, including Low Activity Low Level Waste (LALLW) and Very Low Level Waste (VLLW). It should be noted that the final drum reclassification totals have yet to be established as about 150 difficult to measure drums remain in quarantine pending a more detailed reanalysis.

With more than 95% of ILW-PCM drums reclassified and over 85% of the resulting LLW drums reclassified to lower radiological categories, the LLWR PCM Drum Re-characterisation Project has been transformed into an outstanding success story as a result of the UDASS measurement campaign. Cost savings in excess of nine million pounds (£9M) have been achieved representing a significant return on the investment in employing the ANTECH UDASS measurement service.

Of the total of more than 85% of drums that have been reclassified as below LLW, they can now be sent for disposal to appropriate UK licensed landfill sites thus diverting the waste away from LLWR where disposal space is at a premium. The assessed radiological hazard of the LLWR site has been significantly reduced.

CONCLUSIONS

The UDASS technology has achieved a remarkable success and potentially represents a sea change in the effectiveness of radioactive waste assay technology for drum measurements. Increased measurement sensitivity combined with innovative measurement techniques has provided a method to identify both inhomogeneity in drum matrices and in radioactive source distributions. The UDASS technology incorporates increased measurement sensitivity and significantly improves the effectiveness and accuracy of radioactive waste measurements by correcting for inhomogeneity in the distribution of matrix attenuation (density correction) while at the same time correcting for the radioactive source distribution within a waste drum. The UDASS measurement and drum reclassification technology has the potential to significantly reduce the stockpiles of legacy ILW and TRU waste and reduce radiological inventories worldwide. It can also divert much newly created radioactive waste so that it does not contribute to existing ILW or TRU waste inventory.

UDASS measurements at LLWR have resulted in more than 95% of the ILW drums being reclassified as either LLW or below LLW. These UDASS measurement results have allowed LLWR and Nuclear Waste Services to save most of the costs of ILW disposal. In addition, a total of more than 85% of the LLWR drums have been reclassified as below LLW facilitating their disposal to appropriate UK licensed landfill sites thus diverting the waste away from LLWR where disposal space is at a premium. Aside from cost savings which are significant, the UDASS measurements have significantly reduced the radiological hazard of the LLWR site.

ANTECH continues to develop the UDASS technology. As an example, Figure 10 is a photograph of a UDASS installed in a standard transportable high cube 6 metre (20 ft) ISO shipping container designed for in-field use. This instrument will be deployed to the Los Alamos National Laboratory in early 2023 under US DOE supervision to perform trial measurements of TRU drums on the site.

Fig. 10. UDASS housed in a standard transportable high cube 6 metre (20 ft) ISO shipping container designed for in-field use. This instrument will be deployed under DOE supervision to perform trial measurements of TRU drums at the Los Alamos National Laboratory site.

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