Reclassification of Legacy Wastes to Minimise Disposal Costs and Phantom Inventory – The UDASS Experience at LLWR

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ABSTRACT

The management of nuclear waste has evolved and improved over the decades but remains dependent on accurate characterisation. With the refinement of non-destructive radiometric assay techniques which focus on minimising the measurement uncertainty and hence the reported radioactive inventory, it is possible to revisit the classification of legacy wastes leading to considerable savings in storage and disposal costs. The UK Low Level Waste Repository (LLWR) had 1808 drums of legacy waste from the Magazines that had been classified as ILW PCM (Intermediate Level Waste - Plutonium Contaminated Material) based on historical measurements. A small sample had been re-measured and found to be low level waste, so a campaign was initiated to re-assay all the drums to correctly consign them to the lowest radiological category. The ANTECH UDASS (Universal Drum Assay and Segregation Service), developed from extensive experience in manufacturing waste assay instruments and conducting services, was selected. The UDASS incorporates numerous refinements and updates to existing measurement technologies to optimise measurement efficiency and minimise measurement error. Examples include the variable aperture collimator permitting optimal measurement of a wide range of drum activities and a specialised high efficiency high purity Germanium (HPGe) detector to enhance measurement performance. The UDASS incorporates several operation modes which, after a pre-scan of the waste drum, delivers important information. One can select tailored settings to optimize measurement results as a function of the drum matrix contents heterogeneity. The measurement results are scrutinised by experienced radiometric physicists to ensure that the results are valid prior to their issue and use. The ANTECH UDASS measurement process has been subject to rigorous testing, validation and approval by LLWR and Sellafield technical authorities and accredited to ISO 17025 by the UK Accreditation Service (UKAS). The measurement campaign has resulted in 90% of the drums being reclassified from ILW to Low Level Waste (LLW) and 80% of the resulting LLW were reclassified to categories below LLW permitting landfill disposal. Savings of over £9MGBP and a significantly reduction of the LLWR site radiometric inventory were achieved.

INTRODUCTION

It has been known for some time that significant quantities of higher activity radioactive waste around the world have been overclassified. This is particularly the case in the UK with Intermediate Level Waste with Plutonium Contaminated Material (ILW-PCM) and in the USA with Transuranic Waste (TRU). At the same time large quantities of this assumed higher activity waste have been accumulating, posing a very costly disposal problem in the future.

Waste over classification has arisen through a variety of factors. Typically, radioactive waste characterisation errors are large, often due to the use of inappropriate measurement methods or excessive conservatism in waste measurement assumptions. In the past, conservatism was viewed as increasing safety. This has led to significantly overstating the activity of radioactive waste inventories, in some cases by as much as a factor of 10,000. A major cause has been the often-inappropriate use of Open Detector (OD) based assay methods 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.

The ANTECH UDASS (Universal Drum Assay and Segregation System) has evolved from earlier drum assay instruments and is based on years of experience designing, building, and operating radioactive waste measuring systems at ANTECH (A. N. Technology Ltd.). It has the novel feature that it incorporates several assay technologies in a single instrument platform in order to optimise the waste drum measurement process. 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. The transmission source housing can be seen attached to the pillar on the left and the HPGe high efficiency gamma ray detector is located in the housing attached to the pillar on the right of the photo.

The first of these technologies is the Open Detector (far field) Geometry method [1] and it remains useful for initial screening of drums and to provide an assay result for those drums that can be

shown to be homogeneous. The second method, which is applicable to heterogenous waste drums, is the Wide Range Segmented Gamma Scanner (WR-SGS). The WR-SGS [2] was developed by ANTECH in 2009 as an extension to the capabilities of the original SGS which was developed by Jack Parker, David Jones and Ray Martin [3] at Los Alamos in 1977. A third technology, applicable to highly heterogenous waste drums, is the Tomographic Gamma Scanner (TGS) [4], developed by Robert Estep and others at Los Alamos in 1995.

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. The Open Detector measurement capability for homogeneous drums was added, and the hardware platform was further extended 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 for difficult to measure heterogenous drums.

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). Although the UDASS can be employed as a general-purpose assay instrument for both high and low activity waste in 200 litre drums and 85 US gallon overpack drums, it is primarily focused on reclassifying legacy waste to the lowest relevant radiological category. A more comprehensive description of the UDASS technology and its development is given in reference [5].

UDASS MEASUREMENT METHODS

The UDASS uses a single high efficiency HPGe gamma ray detector that is operated in either Open or Collimated detector mode. It also employs a variable aperture collimator (VAC) that incorporates a tungsten filter for very high dose-rate or high activity measurements. (The filter is nor required for measurements at the waste segregation levels described in this paper). A strong Eu-152 transmission source (typically 10 mCi (370MBq)) housed in a tungsten shield can be seen in Figure 1. When not in use for transmission measurements, the transmission source shield is placed in a shielded safe at the base of the instrument to reduce the background radiation seen by the detector.

Essentially the UDASS divides drums into the two categories of homogeneous (or approximately homogeneous) and heterogenous, in terms of the distributions of both density and activity within the drum matrix. The status of each measured drum is established by the Pre-Scan process, involving both an emission and a transmission Pre-Scan using the WR-SGS method before the main measurement is undertaken. Based on the results of the Pre-Scans, an algorithm within the UDASS software decides if the drum is heterogenous or homogeneous and recommends the appropriate main measurement method to the operator. Relatively few drums meet the criterion for homogeneity and the default condition is to assume that a drum is heterogenous.

Homogeneous Waste Drums

If the drum matrix is assessed as homogeneous in both matrix and source distribution, a variant of the Open Detector measurement, designated the Dynamic Open Detector (DOD) Method is employed for the main measurement. Compared with the WR-SGS method, for appropriate homogeneous drums, the DOD method delivers the lowest counting uncertainty and therefore the best possible measurement result. It represents a significant improvement over conventional OD measurements.

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, it represents a significant improvement over conventional Open Detector measurement technologies. The DOD method is based on helical scanning of the drum with a large Variable Aperture Collimator opening and the emission data acquisition process employs list mode data acquisition.

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 by using a average attenuation factor calculated from the drum weight and not considering density variations within the volume of the drum.

Heterogenous Waste Drums

Heterogenous Waste Drums are measured using the WR-SGS method which has been described extensively elsewhere [2,5]. The method combines the WR-SGS emission scan corrected with the transmission data obtained during the transmission Pre-Scan. All the data is collected using list mode acquisition.

As an additional (and optional) method for improved matrix attenuation correction of highly heterogenous waste drums, TGS transmission has been implemented and the method is designated the TTSGS as it is employed in conjunction with WR-SGS emission measurement data. It also provides visual images of the waste drum attenuation (density) distribution similar to low resolution X-ray images. A disadvantage of this extra measurement stage is that it requires additional measurement time for the separate TTSGS measurement. Image quality and attenuation correction improve as the TTSGS measurement time is extended.

TESTING AND VALIDATION

The design of the UDASS was based on the innovative technology of the ANTECH WR-SGS which has evolved over time. The WR-SGS is a proven assay instrument at technology readiness level (TRL) 9, fully developed and implemented. Like the WR-SGS, the UDASS has been subjected to rigorous testing and in addition, an exceptional level of independent scrutiny and assessment by third parties. The UDASS is also at TRL 9.

Under the terms of a measurement service contract with the UK national Low Level Waste Repository (LLWR), UDASS software processes were examined and assessed, and a team of physicists seconded from Sellafield witnessed and approved both Factory and On-Site Testing of the UDASS, as well as the quality of measurements during the contract. As many of the drum measurement results revealed drums with very low activity, a comprehensive review of UDASS measurement errors was undertaken based on earlier work [6]. UDASS minimum detectable activity (MDA) calculations based on the ISO-11929-3 standard were reviewed and combined with the systematic error data to establish definitive measurement errors for UDASS measurements. The calculations, estimates and assessments have been reviewed and approved by Sellafield staff.

A further independent assessment by the UK Accreditation Service (UKAS) of the UDASS measurements and measurement process including blind test measurements was undertaken. At the conclusion of this assessment, the UDASS measurement process was granted UKAS Accreditation under ISO/IEC:17025 as an in-field laboratory measurement technique.

LLWR MEASUREMENTS OF INTERMEDIATE LEVEL WASTE

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). The contract required the UDASS to measure 1,810 legacy ILW-PCM drums stored on the site from earlier site decommissioning activities, with the aim of reclassifying as many drums as possible. 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 or disposal path except transport to the Sellafield Site for indefinite storage.

The UDASS was positioned inside the ILW drum store at LLWR where the drums were kept. Fig. 2 is a photo of the UDASS which was placed inside a protective cage inside the store.



Fig. 2. UDASS in operation in the ILW store at LLWR.

Both DOD and WR-SGS measurement methods were available for the measurement campaign which began at LLWR in the summer of 2021. Most drums were assayed using the WR-SGS method as relatively few drums passed the UDASS selection criteria for both homogeneity of drum matrix and activity distribution. For DOD measured homogeneous drums, in most cases the low activity was not measured but inferred from MDA values.

Three example drum measurements have been selected to illustrate the UDASS measurement process. For each example the Summary measurement report is presented followed by a plot of the measured activity in Bq for each drum segment and the transmission integral count rate (ICR) in counts per second (cps). Example 1, (Fig. 3a and 3b) is an example of a heterogenous drum containing a discrete solid object. The drum regions have been highlighted and labelled for clarity.

ANTECH						Gam	maScanPro Software
UDASS Report: 3016-ReAnalysis[16	MAY2023_153	34]_SGS					
Summary							
Category	Units	Value	Uncertainty	Level	Test Case Value		Permitted Limit
Gross Mass(*)	kg	61.00	0.50				
Net Mass	kg	40.00	0.50				
Volume	1	2.13E+002	1.00E+000				
LLW Alpha Concentration	Bq/g	7.76E+002	1.55E+001	2σ	7.92E+002	≤	4.00E+003
LLW Beta-Gamma Concentration	Bq/g	2.11E+002	2.80E+000	2σ	2.14E+002	≤	1.20E+004
HV-VLLW Activity Concentration	Bq/g	9.87E+002	1.70E+001	2σ	1.00E+003	>	4.00E+000
LA-LLW Activity Concentration	Bq/g	9.87E+002	1.70E+001	2σ	1.00E+003	>	2.00E+002
VLLW Activity Concentration	Bq/cm^3	2.82E+002	4.38E+000	2σ	2.87E+002	>	4.00E+000
Total Activity	Bq	6.02E+007	8.42E+005	2σ			
Total Pu Mass	g	0.02	0.00	2σ			
U-235 Mass	g	0.00	0.00	2σ			
Total Fissile Mass	g	0.02	0.00	2σ			

Fig. 3a. Example 1. Measurement results table for an example heterogenous matrix drum which contained a solid object measured using the WR-SGS method.

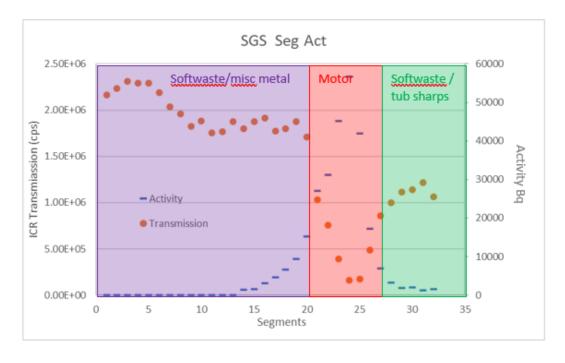


Fig. 3b. Example 1. Plot of activity (Bq) and transmission ICR (cps) for an example heterogenous matrix drum which contained a solid object measured using the WR-SGS method.

Example 2, (Fig. 4a and 4b) is an example of a drum which contained a HEPA filter. Although drums containing HEPA filters do not pass the UDASS homogeneity criteria, they have also been successfully measured using the DOD measurement method.

ANTECH						Gam	maScanPro Software
UDASS Report: 3072-ReAnalysis[17]	NOV2021_125	4]_SGS					
Summary							
Category	Units	Value	Uncertainty	Level	Test Case Value		Permitted Limit
Gross Mass(*)	kg	42.00	0.50				
Net Mass	kg	21.00	0.50				
Volume	1	2.13E+002	1.00E+000				
LLW Alpha Concentration	Bq/g	6.97E+002	1.39E+001	2σ	7.11E+002	≤	4.00E+003
LLW Beta-Gamma Concentration	Bq/g	3.30E+002	5.25E+000	2σ	3.35E+002	≤	1.20E+004
HV-VLLW Activity Concentration	Bq/g	1.03E+003	1.86E+001	2σ	1.05E+003	>	4.00E+000
LA-LLW Activity Concentration	Bq/g	1.03E+003	1.86E+001	2σ	1.05E+003	>	2.00E+002
VLLW Activity Concentration	Bq/cm^3	2.02E+002	2.81E+000	2σ	2.05E+002	>	4.00E+000
Total Activity	Bq	4.31E+007	5.31E+005	2σ			
Total Pu Mass	g	0.01	0.00	2σ			
U-235 Mass	g	0.81	0.01	2σ			
Total Fissile Mass	g	0.82	0.01	2σ			

Fig. 4a. Example 2. Measurement results table for an example matrix drum which contained a HEPA filter measured using the WR-SGS method.

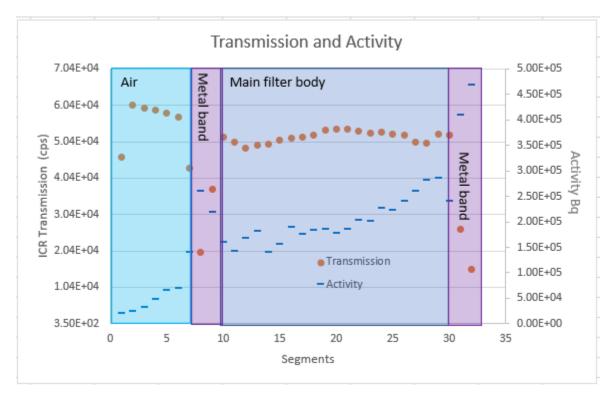


Fig. 4b. Example 2. Plot of activity (Bq) and transmission ICR (cps) for an example matrix drum which contained a HEPA filter measured using the WR-SGS method.

Example 3, (Fig. 5a and 5b) is an example of a drum which contains a matrix of soft waste. Although this drum failed the homogeneity test, other soft waste drum were measured using the DOD method.

ANTECH						Gam	maScanPro Software
UDASS Report: 1450-ReAnalysis[19.	JAN2023_0846	SGS					
Summary							
Category	Units	Value	Uncertainty	Level	Test Case Value		Permitted Limit
Gross Mass(*)	kg	54.00	0.50				
Net Mass	kg	33.00	0.50				
Volume	1	2.13E+002	1.00E+000				
LLW Alpha Concentration	Bq/g	2.10E+002	2.16E+001	2σ	2.31E+002	<	4.00E+003
LLW Beta-Gamma Concentration	Bq/g	1.12E+002	6.29E+000	2σ	1.19E+002	≤	1.20E+004
HV-VLLW Activity Concentration	Bq/g	3.22E+002	2.61E+001	2σ	3.48E+002	>	4.00E+000
LA-LLW Activity Concentration	Bq/g	3.22E+002	2.61E+001	2σ	3.48E+002	>	2.00E+002
VLLW Activity Concentration	Bq/cm^3	8.14E+001	6.51E+000	2σ	8.79E+001	>	4.00E+000
Total Activity	Bq	1.74E+007	1.37E+006	2σ			
Total Pu Mass	g	0.00	0.00	2σ			
U-235 Mass	g	0.31	0.03	2σ			
Total Fissile Mass	g	0.32	0.03	2σ			

Fig. 5a. Example 3. Measurement results table for an example soft waste matrix drum measured using the WR-SGS method.

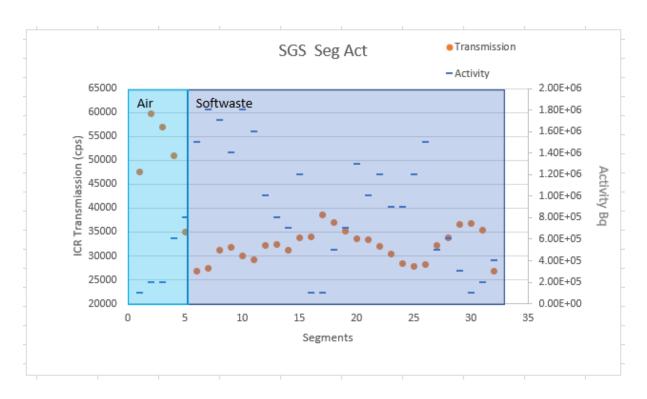


Fig. 5b. Example 3. Plot of activity (Bq) and transmission ICR (cps) for an example soft waste matrix drum measured using the WR-SGS method.

The drum in Example 1 is an interesting case in that the contents were relatively well described as confirmed by the transmission plot. It contains a "discrete item", in the form of a motor with relatively higher Am-241 activity. Such discrete items are of specific interest to LLWR who wish to assign both mass and activity to them. With the UDASS emission and transmission data and plots, this is possible to achieve with smaller errors than was possible before. All of the drums described in the three examples were reclassified from ILW-PCM to LLW.

RESULTS OF THE LLWR PCM DRUM RE-CHARACTERISATION PROJECT

Employing the UDASS for the PCM Drum Re-characterisation Project at LLWR has been an outstanding success. Over 90% of the 1,810 ILW-PCM drums have been reclassified to LLW or lower categories and of those resulting LLW drums, more than 85% have been reclassified to below LLW, mostly to VLLW. Figure 6 shows the revised distribution of drum radiological categories as a result of the drum reclassification following the UDASS measurements of ILW-PCM drums at LLWR.

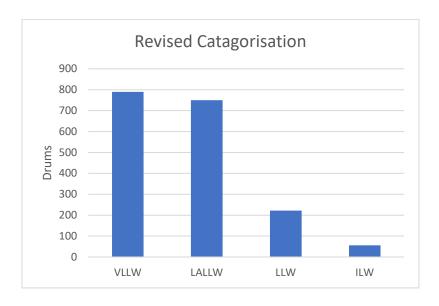


Fig. 6. Number of waste drums plotted as a function of revised radiological category following UDASS drum reclassification.

Both Low Activity Low Level Waste (LALLW) and Very Low Level Waste (VLLW) can be consigned to licenced landfill for disposal.

Figure 7 is a graph (blue) of Frequency versus Specific Activity for UDASS measured values of drum activity with only statistical uncertainty included. The second overlayed graph (beige) is the plot of 'Not Greater Than' activity with systematic uncertainty (2 Sigma) added to the values. Adding this uncertainty shifts the activity towards the higher radiological categories. Note that while not many reclassified drums fall into below 4 Bq/g category of high volume VLLW, many more fall into the other VLLW category of below 4 Bq/cm³.

The graph in Figure 8 displays the Statistical Uncertainty (2 Sigma) in the measured data as a function of Specific Activity in Bq/g.

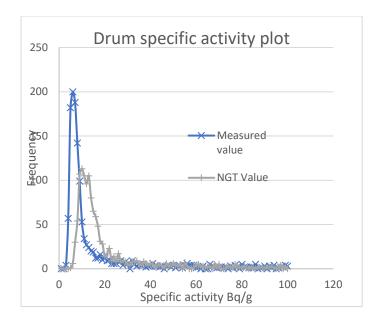


Fig. 7. Frequency versus Specific Activity: UDASS measured values of drum activity with only statistical uncertainty and 'Not Greater Than' values with systematic uncertainty (2 Sigma) added.

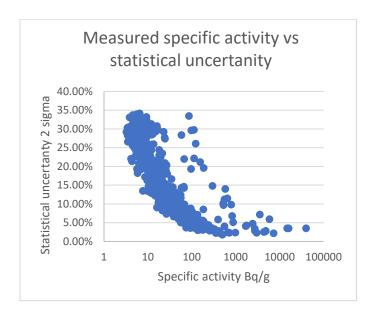


Fig. 8. Statistical Uncertainty (2 Sigma) in the measured data as a function of Specific Activity in Bq/g

CONCLUSIONS

As a result of employing the UDASS for the PCM Drum Re-characterisation Project at LLWR, over 90% of the 1,810 ILW-PCM drums have been reclassified to LLW or below. This has resulted in savings in waste disposal costs of over £9M for the 1,810 drums measured at LLWR [5, 6, 7] and significantly reduced the site radiological hazard by more than a factor of 10. Even with a conservative assumed reclassification rate of 40%, the UDASS has the potential to reclassify the vast quantities of overclassified higher activity waste saving space in repositories and waste stores, reducing both radiological inventories and radiological hazards.



Fig. 9. UDASS housed in an ISO Container for Transport and On-Site Deployment. The system in the photo is being shipped to the Los Alamos National Laboratory (LANL) in the US as part of a US Department of Energy trial project to reclassify TRU drums at LANL.

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