



JRC TECHNICAL REPORT

Good Practice Guide for Validation of a Waste Characterisation System for Very Low, Low and Intermediate Level Radioactive Waste

*16ENV09 MetroDECOM II,
Deliverable D4*

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2021



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EU Science Hub
<https://ec.europa.eu/jrc>

JRC123784

EUR 30761

PDF

ISBN 978-92-76-40051-6

ISSN 1831-9424

doi:10.2760/748464

Luxembourg: Publications Office of the European Union, 2021

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How to cite this report: De Felice, P., Bogucarska, T., Raiola, F., Pedersen, B., *Good Practice Guide for Validation of a Waste Characterisation System for Very Low, Low and Intermediate Level Radioactive Waste*, EUR 30761, Publications Office of the European Union, Luxembourg, 2021, ISBN 978-92-76-40051-6, doi:10.2760/748464, JRC123784.

16ENV09 METRODECOM II

DELIVERABLE D4: GOOD PRACTICE GUIDE, WP3 VALIDATION O WASTE CHARACTERISATION SYSTEM FOR VERY LOW, LOW AND INTERMEDIATE LEVEL RADIOACTIVE WASTE

**LEAD PARTNER: JRC
PARTNER: ENEA**

Due date: February 2021

Approved by consortium date: 16 March 2021

Submission date: 22 February 2021

This project has received funding from the EMPIR programme co-financed by the Participating States and from the European Union's Horizon 2020 research and innovation programme.

Good practice guide

D4: Good practice guide for validation of a Waste Characterisation System for very low, low and intermediate level radioactive waste

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ABSTRACT

The present report describes the lessons learned from the taking into operation of the two main measurement stations of the Waste Characterisation System (WCS). This includes the SGS/TGS gamma station and the passive/active neutron station.

The report describes details of the detection systems, the multiple detailed investigations undertaken to determine their assay characteristics, and finally the observed performance values determined by assay methods applying known radioactive and nuclear material standards. The work carried out at the two measurement stations was organised according to a formal validation procedure, outlined in the report, for the applied assay methods. The multitude of investigations needed to comply with the formal validation procedure, could not be completed in the limited time of the present project. The investigations undertaken, however, are a useful insight into technical issues and methodologies to be addressed in any validation process of assay methods of radioactive and nuclear materials. The achieved assay results provide useful target values for others about to embark on validation of methods for radioactive waste characterisation.

GLOSSARY/ABBREVIATIONS

| | |
|--------|--|
| CNAPI | Carta Nazionale delle Aree Potenzialmente Idonee (CNAPI) |
| CMT | Cemented waste |
| D&WM | Decommissioning and Waste Management |
| FCC | Facility Control Computer |
| GSC | Gamma Station Computer |
| IAEA | International Atomic Energy Agency |
| ILW | Intermediate-Level Waste |
| ISF | Interim Storage Facility |
| ISOCS | In-Situ Object Counting System |
| ITSP | Innovative Techniques and Standardization of Procedures |
| LLW | Low-Level Waste |
| MTL | Metallic waste |
| NSC | Neutron Station Computer |
| PLC | Programmable Logic Controller |
| RBL | Rubbles waste |
| ROI | Region of Interest |
| SGS | Segmented Gamma Scanner |
| TC/TCO | Technological Combustible waste |
| TGS | Tomography Gamma Scanner |
| TNC | Technological non-combustible waste |
| TSGS | Tomography/Segmented Gamma Scanner |
| VLLW | Very Low-Level Waste |
| WBBC | Weight Barcode Bridge Computer |
| WCF | Waste Characterisation Facility |
| WCS | Waste Characterisation System |

EXECUTIVE SUMMARY

The present report constitutes the Good Practice Guide resulting from the work carried out under the Work Package 3 of the project MetroDECOM II. The task was to take into operation the two major measurement stations at the WCS facility of JRC, the passive/active neutron station, and the SGS/TGS gamma station. The approach to this task was, if not to complete all measurement objectives during the limited time of this project, to follow a scheme of validation of the assay methods applied at the two measurement stations. For this purpose, Chapter 7 outlines the formal requirements in a validation procedure of non-destructive assay methods applied to radioactive and nuclear materials. The intention is that this procedure, the numerous instrument qualification tasks undertaken, and the measurement campaigns on simulated and real waste reported in the Chapters 10 to 14, can serve as a guide to others with the formidable task of developing a validation procedure for a specific non-destructive assay instrumentation or method.

For both the SGS/TGS gamma station and the passive neutron station, the instrumentation is described in detail together with the characteristics of the radiation detection systems, and a summary of the analytical methods. From there follows discussions of a series of experiments on simulated waste drums composed of the various real waste matrices in drums where radiation standard sources can be inserted. The measurement campaigns also included assay of 12 real waste drums.

Finally, this report gives a short description of lessons learned, during the 42 months project, related to taking into operation of a waste characterisation system of low and intermediate level waste.

At the start of the project, the intention had been to include the active neutron station in this work. However due the general Covid situation in 2020, the experimental work for this part could not be completed and was removed from the objective. In spite of this, the present report is a significant contribution to metrology for radioactive and nuclear waste as a collective of all the steps necessary in the validation process of any new waste characterisation system.

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1 INTRODUCTION

The variety of non-destructive assay techniques are used within the nuclear industry to provide information of the radionuclide inventory of packages containing radioactive waste. The present report (Good Practice Guide (GPG)) shows the validation approach of the Waste Characterisation System (WCS) for very low, low and intermediate level radioactive waste applying the gamma spectroscopy and passive neutron counting methods. The WCS is in operation at the JRC Ispra site of the European Commission. The validation activities performed and described in this GPG have been developed in the frame of the 16ENV09 MetroDECOM II project funded by the European Metrology Programme for Innovation and Research (EMPIR). Wherever possible validation activities have been performed in line with the requirements of the ISO17025:2017 standard.

2 THE NUCLEAR WASTE ISSUE

2.1 ORIGIN OF WASTES

Globally, significant amount of radioactive waste is generated from a broad range of activities involving the operation and decommissioning of nuclear facilities, activities using radioisotopes in science, industry and medicine, including disused sealed radioactive sources. Such waste needs to be managed in a way that keeps people and the environment safe over long periods.

2.2 LEGISLATIVE REQUIREMENTS

In Italy (the country where the activity related to the present report is carried out), medium and high activity waste (that is, those which lose radioactivity in thousands of years and which, in order to be definitively storage, require the availability of a geological repository) will be temporarily stored in the National Repository. Very short-lived radioactive waste, which after losing its residual radioactivity can be disposed as conventional waste (no longer radioactive), is excluded. The company Sogin S.p.A is the responsible legal subject of localization, construction and use of the National Depository of radioactive waste and Technological Park.

Currently, a national map of potentially suitable areas, (Carta Nazionale delle Aree Potenzialmente Idonee) CNAPI, has been published (ex D.Lgs. n. 31/2010) since 5 January 2021 (67 proposed sites), with the mandate to investigate suitability of the identified areas, based on socio-environmental and technical characteristics. The preliminary project, and related documentation, has been published and can be consulted on the website www.depositonazionale.it.

In accordance with International Atomic Energy Agency (IAEA) standards [1], the Ministerial Decree 7 August 2015 “Classificazione dei rifiuti radioattivi, ai sensi dell’Art. 5 del decreto legislativo 4 marzo 2014 n. 45” (Art.5 of D.Lgs. n. 45 of 4th March 2014) [2] has reviewed the national classification of radioactive wastes establishing the following categories:

1.1.1 VLLW-Very Low Level Waste;

1.1.2 LLW-Low Level Waste);

1.1.3 ILW-Intermediate Level Waste;

1.1.4 HLW-High Level Waste;

and correspondingly assigning final destination storage facilities as summarised in Table 1.

Table 1 - Categories and their characteristics as described in the Art.5 of D.Lgs. n. 45 of 4th March 2014 for the radioactive waste at the JRC-Ispra [2].

| Category | Condition and/or Activity concentrations | | Final destination |
|--------------------------------|--|---|---|
| Exempted | Art. 154 comma 2 of D.Lgs. n. 230/1995 Art. 30 or art. 154 comma 3-bis of D.Lgs. n. 230/1995 | | In compliance with the disposition of D.Lgs. n. 152/2006 |
| Very short lived waste | $T_{1/2} < 100$ d Achieving into 5 years these conditions: Art. 154 comma 2 of D.Lgs. n. 230/1995 Art. 30 or art. 154 comma 3-bis of D.Lgs. n. 230/1995 | | Temporary storage (art.33 D.Lgs. n. 230/1995) and disposal in agreement with the dispositions (D.Lgs. n.152/2006) |
| Very low activity waste (VLLW) | ≤ 100 Bq/g (in which Alpha ≤ 10 Bq/g) | Reaching in $T \leq 10$ years of this condition: Art. 30 or art. 154 comma 3-bis of D.Lgs. n. 230/1995 | |
| Low activity waste (LLW) | Radionuclides short lived ≤ 5 MBq/g Ni-59, Ni-63 ≤ 40 kBq/g radionuclides long lived ≤ 400 Bq/g | Not reaching in $T \leq 10$ years of this condition: Art. 30 or art. 154 comma 3-bis of D.Lgs. n. 230/1995 | Surface storage facility, or at low depth, with engineering barriers (National Storage D.Lgs. n. 31/2010) |
| Medium activity (ILW) | Short lived radionuclides > 5 MBq/g Ni-59, Ni-63 > 40 Bq/g long lived radionuclides > 400 Bq/g no heat released | Alpha emitter radionuclides ≤ 400 Bq/g and beta-gamma nuclides in concentration that respect the radioprotection limits established for the superficial storage facility Radionuclides in concentration that do not respect the radioprotection limits established for the superficial storage facility | Temporary storage facility of the National Storage (D.Lgs. n.31/2010) for the final geological disposal |
| High activity (HLW) | Released of heat or high concentration level of long lived radionuclides, or both conditions | | |

2.3 GAMMA AND NEUTRON MEASUREMENTS

In order to determine a waste category, suitable measurement methods are needed that can be performed on materials properly packed in special waste containers.

Measurement methods range from gamma spectrometry (able to perform photon emitters identification and quantification) to passive and active neutron measurements (needed to quantify spontaneous fission and fissile material). Some methods also rely on tomography techniques in order to get information about the distribution of the radionuclides inside each container. Additional dosimetry measurements can be performed on the external part of each

container. In some cases (low energy photon emitters and pure beta emitters), gamma and neutron measurements need to be complemented with radiochemical methods. These last methods will not be covered by the present document.

On JRC Ispra site standard drums with 220 L and 440 L internal volume have been selected and are used as waste containers. As described in the next chapters, a fully automatized WCS has been developed for waste characterisation based on gamma and neutron measurements supported with tomography and dosimetry measurements. It is then essential and mandatory to have a validated method to perform this characterisation of the waste. While gamma measurement is the basic tool for waste characterisation, as most of the radionuclides emit photons with energy higher than 40-50 keV with photon intensities suitable to provide acceptable detection limits, the need of additional neutron measurements is documented in the following paragraph.

2.3.1 Specific needs for neutron measurements

Neutron measurements of waste packages serve the single purpose of quantifying the fissile material constituents of the waste. The reasons for paying special attention to fissile materials, are related to the high specific alpha activity of most actinides, and the special interest to ^{235}U and Pu isotopes under international nuclear safeguards.

Some specific circumstances, among others, that make mass assay of the two major actinides in waste of particular importance, are:

- 1 Waste repositories operate with waste acceptance criteria derived from their license limitations. Often waste packages are classified according to activity level per gram of waste, and type of radiation. In case of waste containing actinides, the alpha activity must be taken into account. The content of long-lived alpha emitters (half-life greater than 5 years) often decides the classification of the waste due to their tight acceptance criteria. The type of actinide present in the waste matters enormously. E.g., the specific activity (alpha activity per gram of material) of a Pu sample is typically six order of magnitude higher than for a U sample. In other words, if the waste item contains Pu, it is essential to quantify this element for the correct categorisation of the waste item.
- 2 The two major actinides (U and Pu) require special attention because safeguards control requirements of organisations such as IAEA and Euratom. For example, the IAEA has recently developed a guideline that quantifies the maximum levels of ^{235}U or Pu in different types of waste package for accepting their removal from the facility material balance [3]. Only in case of demonstrably small quantities fissile material, or if the fissile constituents are so diluted that for all practical purposes they cannot be recovered, can the facility operator remove the waste item from safeguards control.

The passive/active neutron station of WCS implements both passive measurements (to quantify the even mass-number Pu isotopes) and active measurements (to quantify either ^{235}U , or small masses of either Pu or ^{235}U).

The purpose of the passive neutron assay is to quantify spontaneous fissile isotopes present in the waste. This is useful when, for example, the gamma scanning measurement has identified the presence of Pu in the waste. Even mass-number Pu isotopes undergo spontaneous fission that allows determining their mass by means of the so-called neutron correlation analysis. In this method, the measurement of the signal multiplets allows the determination of the spontaneous fission rate and with that the Pu mass. In these cases, the passive neutron assay yields a more accurate determination of the Pu mass when compared to other methods. The reasons are that neutrons, in contrast to gamma-rays, penetrate relatively easy high-density matrices such as metal, and that the neutron correlation technique allows to determine the neutron counting efficiency together with the fissile mass. In the present work, measurements were performed on five types of simulated waste matrices in 220 L waste drums with calibrated sealed PuO_2 standards inserted.

2.4 APPROACHES

Given the socio-economic importance of waste assay methods, reliable, accurate and validated (§7) procedures are needed. In this context, standard methods are of great support and represent the basic platform for other user-developed methods, eventually more focused on specific scopes.

The basic normative methods are reported in the following list:

1. ISO 20042:2019 Measurement of radioactivity — Gamma-ray emitting radionuclides — Generic test method using gamma-ray spectrometry [4].
2. ISO 19017:2015 Guidance for gamma spectrometry measurement of radioactive waste [5].
3. ISO 8529-1:2001 Reference neutron radiations -- Part 1: Characteristics and methods of production [6].
4. ASTM, Standard Test Method for Nondestructive Assay of Special Nuclear Material in Low-Density Scrap and Waste by Segmented Passive Gamma-Ray Scanning, West Conshohocken, PA: ASTM International, 2018 [7].

A further useful document is the following:

5. D.M. Cifarelli and W. Hage, "Models for a three-parameter analysis of neutron signal correlation measurements for fissile material assay", Nuclear Instruments and Methods in Physics Research A251 (1986) 550 – 563 [8].

3 EMPIR 16ENV09 MetroDECOM II project

3.1 PROJECT OBJECTIVE

This project addresses one of the most significant environmental challenges facing EU member states: ensuring the safe disposal of radioactive waste from decommissioning nuclear sites. The key to dealing with such wastes is quantifying the radioactivity content, so that decommissioning can be planned and implemented minimising the risk to members of the public and the environment

The first generation of nuclear power plants and reprocessing facilities is coming to the end of their working lives. 91 power plants are being decommissioned in the EU; most of the remaining 129 reactors plus fuel cycle facilities will also be in decommissioning by 2030.

The aim of the decommissioning process is to clear the site, while minimising the risk to the public and the environment from the hazardous waste arising. The cost of decommissioning and waste management in the EU is estimated to be in excess of 150 billion Euro.

The key to safe and cost-effective disposal of the waste is accurate characterisation – determining the physical, chemical, and radiological characteristics of the material. This enables nuclear site operators to plan the demolition process, assign the waste to the most cost effective disposal route and then to monitor that the waste is being stored safely. The metrological challenge is the disposition and quantities of hazardous materials are not well known. There has been significant progress towards developing the new techniques needed in EMRP JRP ENV09 MetroRWM and ENV54 MetroDecom; this project focusses on bringing the techniques into use on nuclear sites and developing further innovative solutions based on lessons learned.

Regulatory bodies and international organisations have therefore carried out detailed studies of technical needs in the field. The common themes that have been raised are:

- 1 improvements in capability;
- 2 harmonisation and quality assurance;
- 3 sharing knowledge.

The improvements in capability that are required include rapid, on site measurements, improving the accuracy and traceability of measurements of waste packages.

These needs are reflected in EU Council Directive 2011/70/EURATOM which aims to encourage technical co-operation to improve safe management of radioactive waste and highlights the importance of building public trust and confidence.

EMPIR 16ENV09 MetroDECOM II project divided to five work packages and each one of them covers the following objectives:

1. Work Package 1: To develop in situ methods for the rapid radionuclide characterisation of the different types of materials present on decommissioning sites. This includes the development of novel measurement techniques that improve the mapping of contamination inside nuclear facilities and the determination of statistically valid and effective sampling methods.
2. Work Package 2: To develop and implement traceable measurement systems and methods for waste pre-selection and free release to the environment. This includes the on-site validation of existing waste pre-selection and free release measurement systems and evidence of their traceability to primary standards.
3. Work Package 3: To develop a validated waste repository acceptance characterisation system for use on site with very low and intermediate level radioactive waste (LLW and ILW). The system will include gamma scanning and passive neutron measurements, and will be sensitive to fissile material.
4. Work Package 4: To develop improved measurement systems and methods for monitoring radioactive waste repositories. This includes miniaturised, portable and rapidly deployable gas and water monitoring systems, improved water monitoring systems and systems for monitoring the temperature and strain of inside nuclear waste repositories for the long term. All the systems will be verified with on-site testing.
5. Work Package 5: To facilitate the take up of the technology and measurement infrastructure developed in the project by nuclear decommissioning operators, measurement device producers, radiation protection regulators and standards developing organisations. In addition, to establish in collaboration with the end-user community a European network that will co-ordinate a measurement infrastructure for decommissioning nuclear facilities.

The overall objective of this project is to establish a measurement infrastructure that is accepted and implemented by the nuclear industry for the measurement of radioactivity, temperature and stress needed for nuclear decommissioning [9].

3.2 WP3 DESCRIPTION

The specific objective of Work Package 3 concerns the validation of a WCS for very low, low and intermediate level radioactive waste. The aim of this Work Package is to validate a new WCS for VLLW, LLW and ILW because radionuclide activities are typically higher than measured in the free release system in WP2; this Work Package also includes the non-destructive measurement of fissile materials.

The WCS includes two measurement stations composed of a segmented/tomographic gamma-scanning device and a passive neutron device. The measurement stations are part of a newly refurbished industrial waste handling facility, called WCS, to be operated on an active nuclear decommissioning site. Measurement campaigns on both real and simulated waste containers will be carried out and the performance of the systems in terms of detection limits, accuracy and measurement time will be determined, based on actual measurements.

Task 3.1 covers characterisation of gamma segmented / tomographic measurements, Task 3.2 covers passive neutron measurements and Task 3.3 covers a measurement campaign to demonstrate the robustness of the measurements. The target uncertainties for measurements for both gamma and neutron measurements is 20% for typical decommissioning matrices and a measurement time of 2000 s.

This work package results in a good practice guide on the use of a waste acceptance measurement system using gamma and passive neutron detectors (Deliverable D4), taking into account lessons learned during the measurement campaign using real waste packages.

3.2.1 Task 3.1: Characterisation of a segmented/tomographic gamma scanning device

The aim of this task (Table 2) is to characterise the newly modified JRC waste drum gamma measurement station in terms of characteristic parameters for measurement of standard 220 L waste drums. The measurement sequences and the analysis method will be developed. The measurement campaign will make use of simulated waste matrices and both radioactive and nuclear sealed sources of well-known content in order to determine reliable performance values in preparation for the measurement campaign using real VLLW, LLW and ILW packages in Task 3.3.

Table 2 - Task 3.1: Characterisation of a segmented/tomographic gamma scanning device

| Activity number | Activity description | Partners (Lead in bold) |
|-----------------|--|-------------------------|
| A3.1.1 | JRC will write and ENEA will review a detailed description and characteristics of a SGS/TGS system for 220 litre waste drums. The document will include the technical system, proposed outline measurement methodology. | JRC, ENEA |
| A3.1.2 | JRC will develop and ENEA will review detailed analysis methods, based on the outline documented in A3.1.1. This work includes detailed development of the analysis methods to be applied for the 220 litre drums of both real and simulated waste, and describes the sequence of measurements needed for each waste item. Also included is a description of the measurement report template and the data storage system | JRC, ENEA |
| A3.1.3 | Using input from A3.1.1 and A3.1.2, JRC and ENEA will organise and carry out a measurement campaign at JRC (Ispra) of four months duration on 220 litre simulated waste using well-characterised radioactive and nuclear standards, and standard containers of different matrix materials including metal, concrete, and low-density process waste. The waste containers have re-entrant tubes for implementing a variety of source configurations. The campaign will concentrate on both standard and particularly complicated source configurations in order to estimate the performance range of the instrumentation. | JRC, ENEA |
| A3.1.4 | ENEA will determine the performance values of the instrumentation based on the measurement data from the campaign of simulated waste, including achieved measurement accuracy, detection limits in various matrices (target 0.1 Bq/g for ^{137}Cs in a 60 kg matrix with a measurement time of 2000 s) and measurement uncertainty (target uncertainty 20 %) JRC will review and comment on the results. | ENEA, JRC |

3.2.2 Task 3.2: Characterisation of a passive neutron measurement station

The aim of this task (**Error! Reference source not found.**) is to investigate the performance of a passive neutron measurement station for the characterisation of actinide-containing waste.

Table 3 - Task 3.2: Characterisation of a passive neutron measurement station

| Activity number | Activity description | Partners (Lead in bold) |
|------------------------|--|------------------------------------|
| A3.2.1 | JRC will write and ENEA will review a detailed description and characteristics of a passive neutron detection system for 220 litre waste drums. The document will include the technical system, proposed outline measurement methodology. | JRC, ENEA |
| A3.2.2 | JRC will develop and ENEA will review detailed analysis methods, based on the outline documented in A3.2.1. This work includes developing the analysis methods to be applied for the 220 litre drums of both real or simulated waste, and describing the sequence of active and passive measurements needed for each waste item, including an investigation of how best to take into account the neutron flux attenuation within absorbing matrices. Also included is a description of the measurement report template and the data storage system. | JRC, ENEA |
| A3.2.3 | Using input from A3.2.1 and A3.2.2, JRC and ENEA will organise and carry out a measurement campaign at JRC (Ispra) of four months duration on 220 litre simulated waste using well-characterised nuclear standards, and standard containers of different matrix materials including metal, concrete, and low-density process waste. The waste containers have re-entrant tubes for implementing a variety of source configurations. The campaign will make use of plutonium sources on a variety of source configurations in order to estimate the performance of the instrumentation. | JRC, ENEA |
| A3.2.4 | JRC will determine the performance values of the instrumentation based on the measurement data from the campaign of simulated waste, including achieved measurement accuracy, detection limits in various matrices ((target 0.1 Bq/g in a 60 kg matrix with a measurement time of 2000 s) and measurement uncertainty (target uncertainty 20 %). ENEA will review the results. | JRC, ENEA |

The presence of fissile materials in the waste requires an estimate of the alpha content of the waste item, as well as the fissile material content of the waste to be reporting to nuclear safeguards authorities. This task intends to investigate the performance that can be achieved in an industrial-size system for producing declarations of fissile material content in standard waste containers.

3.2.3 Task 3.3: Measurement campaign of VLLW, LLW and ILW on the JRC decommissioning site.

The purpose of this task (Table 4) is to conduct a measurement campaign using real decommissioning waste using the JRC SGS/TGS system and the JRC passive neutron detection system, and to report the results in standard operational mode. The aim of this task is to produce good practice guides for both gamma measurements of radioactive waste and neutron measurements of fissile-material containing waste.

Table 4 - Task 3.3: Measurement campaign of VLLW, LLW and ILW on the JRC decommissioning site.

| Activity number | Activity description | Partners (Lead in bold) |
|-----------------|--|-------------------------|
| A3.3.1 | JRC will organise and carry out a measurement campaign of real waste items, which will be carried out using the SGS/TGS system for 220 litre waste drums, including data analysis based on the methods implemented under A3.1.2 for gamma-emitting radionuclides. The campaign will be undertaken at JRC Ispra for two months and will concentrate on waste items already in the JRC inventory. A minimum of 12 waste packages will be measured. | JRC |
| A3.3.2 | JRC will carry out and ENEA will review data analysis and reporting of results based on the campaign of real waste from the decommissioning project on the JRC Ispra site (A3.3.1). JRC and ENEA will review the results and document any lessons learned for inclusion in the Good Practice Guide (D4). | JRC, ENEA |
| A3.3.3 | JRC will organise and carry out a measurement campaign of real waste containing fissile materials using the JRC passive neutron detection system, including data analysis based on the methods implemented under Task 3.2 (A3.2.2). The campaign will be undertaken at JRC Ispra for two months and will concentrate on waste items already in the JRC inventory. | JRC, ENEA |
| A3.3.4 | JRC will carry out and ENEA will review data analysis and reporting of results based on the campaign of real waste from the decommissioning project on the JRC Ispra site (A3.3.3). JRC and ENEA will review the results and document any lessons learned for inclusion in the Good Practice Guide (D4). | JRC, ENEA |

3.3 PROJECT PLANNING

Figure 1 illustrates the schedule of Work Package 3.

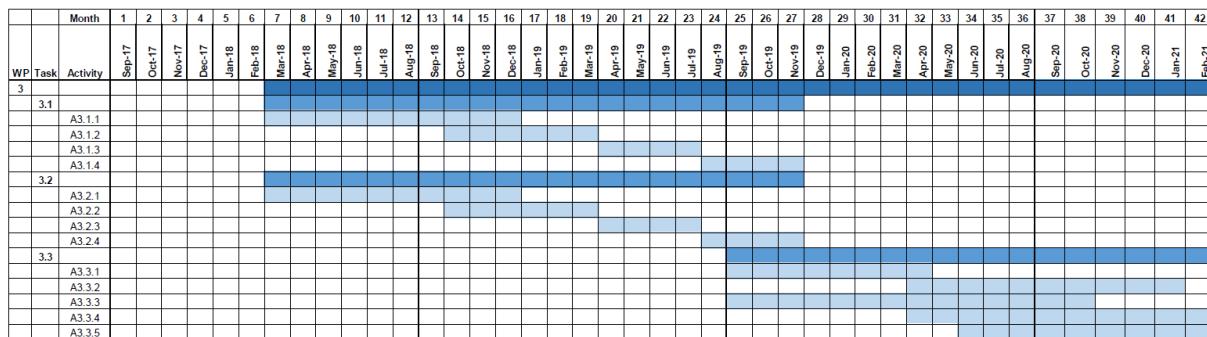


Figure 1 - Gantt chart

3.4 DELIVERABLE D4

JRC and ENEA wrote a Good Practice Guide based on the results from A3.3.2 and A3.3.4. Within the project, the Good Practice Guide refers to the deliverable D4.

The Good Practice Guide represents the lessons learned in undertaking metrological characterisation using the WCS for use on site with very low, low and intermediate level radioactive waste. This includes work on gamma scanning and passive neutron measurements, and sensitivity to fissile material'.

4 EC NUCLEAR D&WM Programme

The European Commission is committed to protect the public and the environment from undue radiological hazards. It has therefore developed a Decommissioning & Waste Management (D&WM) Programme for all nuclear sites of the Joint Research Centre (JRC).

The European Parliament, during its debates on the future EURATOM research programme, requested explicitly that: "JRC builds upon its experience with the decommissioning of JRC

nuclear facilities and further reinforces its research to support safe decommissioning in Europe ". Reinforced in a roundtable meeting between industry and science representative in Brussels in 2012, for future prospects and priorities for European nuclear decommissioning. Besides, in the opening remarks of a meeting organised by JRC in Brussels on 23 January 2018 with representatives of the European Commission, industry and nuclear scientific experts, Vladimír Šucha, ex. JRC Director-General, said: "Nuclear decommissioning is an important emerging field in the EU and internationally, creating opportunities for high-skilled workers and posing new technological challenges. The JRC regularly interacts with industry, research and training organisations to ensure a common understanding of the challenges and promote new research and development initiatives in the field." (see <https://ec.europa.eu/jrc/en/news/supporting-european-expertise-nuclear-decommissioning>). JRC implements several institutional and competitive projects in the field of nuclear decommissioning, waste management and environment remediation. These projects regard not only Research & Development (R&D) but also Education and Training (E&T) activities are either fully funded by the institutional budget or through participation to competitive projects, which are monitored by EC DG RTD or international consortia. EC priorities in this field is to enhance capacity building by reinforcing human capital, to improve standardization for safe decommissioning and waste management and finally to prepare for the vast emerging decommissioning market, which is about to grow drastically considering the aging of the nuclear fleet.

5 ONGOING NUCLEAR D&WM PROGRAMME IN JRC-ISPRA SITE

5.1 HISTORICAL LIABILITIES

During the last decades, most nuclear research activities have been terminated in JRC-Ispra site (Italy). The few exceptions of the remaining activities regard research on NDA techniques for high performance nuclear safeguards, security and waste management. In addition to these ongoing research activities, an important programme on various trainings in the nuclear field is ongoing as well.

The Centre's Historical Liabilities (i.e. those nuclear R&D facilities and radioactive waste management installations that have no future role in supporting the mission of the JRC) include seven large facilities (Figure 2) such as two research reactors (ESSOR and ISPRA 1), a cyclotron, an amount of obsolete irradiated and non-irradiated nuclear materials, and finally various sorts of Low Level Waste (LLW) and Intermediate Level Waste (ILW).



Figure 2 - The seven large facilities at the JRC-Ispra site with two research reactors (top and bottom at the far right side) and a cyclotron (bottom left side).

Some major nuclear installations are placed in the state of "safe conservation" in view of their eminent decommissioning.

5.2 JRC-ISPRA D&WM PROGRAMME

The D&WM Programme in JRC-Ispra started in 1999, managed by the Nuclear Safety and Security Directorate.

The D&WM Programme is highly dependent on the Italian regulatory framework, in particular for the licensing of the decommissioning activities. JRC-Ispra radioactive waste will have to be disposed in a National centralised Repository, that is, those that within 300 years will reach a level of radioactivity that no longer represents a risk to humans and the environment. JRC agreed on appropriate management criteria for such waste, in order to condition it in adequate form.

The JRC's D&WM Programme aims at the progressive elimination of the JRC-Ispra historical Liabilities. Its main objectives are to decommission the shutdown of nuclear facilities and to manage the resulting waste together with the old waste.

Some specific D&WM Programme objectives have already been reached, while others are ongoing, including:

- Refurbishing or building new infrastructure for waste characterisation, treatment/conditioning and temporarily storage of waste arising from new dismantling operations;
- Retrieval, characterisation, treatment and conditioning of existing old waste;
- Removal, treatment and disposal of unused nuclear materials (nuclear fuel and experimental nuclear materials);
- Clean-up and dismantling of installations allowing unrestricted use of the buildings or even the "green field" option, which comprises the demolishing of buildings and the restoration of the land in its initial state;
- Transfer of waste to an Italian national site for disposal for which the Italian Authorities did not know yet about the location or about when the construction could be kicked-off.



Figure 3 - Interim Storage Facility (ISF) at JRC-Ispra (Source: JRC, 2019).

To reach these objectives, the JRC-Ispra Nuclear D&WM Programme include currently more than 30 major projects and services such as:

- Construction and commissioning of highly technological facilities to accommodate characterisation and conditioning of radioactive waste. An example of such facilities for high performance waste characterisation using NDA (with gamma and neutron measurements) and imaging (with tomographic scans) techniques.
- Evacuation of major parts of the obsolete nuclear materials (uranium, plutonium and disused radiological sources).
- Characterisation of the nuclear installations that have to be decommissioned.
- Construction of a facility for the conditioning and the interim storage of the nuclear waste as shown above in Figure 3.

- Re-conditioning of older historical waste that was stored and or disposed in the existing site waste management station.
- Preparation of decommissioning plans for the installations.
- Evacuation of hazardous materials (e.g. alkaline metals).
- Construction of a grouting facility for the immobilisation in waste containers: the design for the grouting facility is ready and it will be constructed in the place of an old cementation plant which is dismantled.
- Qualification of a 5,2 m³ container for low level waste interim storage, transport and final disposal: finalisation of container qualification for different waste streams is expected for end of 2014.
- Retrieval and temporary storage of experimental rigs containing irradiated nuclear material and some spent fuel: cold test for retrieval and temporary storage in the Transit Safe Area (TSA) is concluded and transfer of rigs is to be planned depending on the regulatory authorisations.
- Middle-term storage of spent fuel is envisaged in dedicated casks, the procurement is ongoing.
- Pre-decommissioning and decommissioning activities of major installations: ongoing are clean-up and dismantling activities within existing licenses of the various installations: reactor facility (FARO), hot cells (LCSR) and liquid effluent treatment facility (STRRL); specific decommissioning plans for most of the major installations are in progress.
- Retrieval and treatment of historical waste: retrieval, characterisation and treatment of approximately 6000 bituminised drums: construction of a waste retrieval plant is planned to begin 2015 and off-site treatment is planned for 2017; the tenders for both retrieval facility and off-site treatment are published.
- Treatment and conditioning activities of existing waste and waste produced through decommissioning activities: sorting, repacking, characterisation and preparation for super-compaction and possible metal melting of existing waste and newly produced waste is in progress, in parallel the clearance plant is used for preparing materials for release.
- Completion of evacuation of obsolete non-irradiated nuclear materials: as nuclear research, the Ispra site is handling a big variety of materials in different physical and chemical forms. The today still remaining materials are well characterised and different options for evacuation are prepared or in evaluation.

The initial decommissioning plan at the JRC-Ispra site has foreseen the shutdown of the last existing installation in 2025, which would lead to the end of decommissioning around 2030. However, this timing became unrealistic and it is possible that the schedule may be postponed by as much as a decade.

The D&WM Programme will be concluded when the Ispra site will be brought to the “green field” status: a condition reached after the decommissioning process where building and land are released free of any radiological constraint with an estimated completion on 2030.

6 JRC WCS

6.1 HISTORICAL DEVELOPMENTS

To support the decommissioning the JRC has constructed on the Ispra site the Intermediate Storage Facility (Figure 3) intended to hold all LLW and ILW radioactive wastes from the site, in conditioned form, until their departure to a future National Deposit.

For the purpose of characterising standard waste packages in terms of radioactive and nuclear content, the A.N Technology Ltd. (UK), i.e. ANTECH, was commissioned by JRC, at the end of 2007, to develop a Waste Characterization System (WCS). The WCS was intended for measurement of fission products, uranium and plutonium in waste arising from past, on-going, as well as future nuclear activities at the JRC-Ispra site.

The WCS was designed to analyse 220 L and 440 L drums and to produce assays that include surface dose rate measurement, Tomography Gamma-ray Scanning (TGS), Segmented Gamma-ray Scanning (SGS) and active and passive neutron assay.

WCS was completed in 2012 but was never taken into full operation. WCS was purchased as a turnkey installation, complete with proprietary algorithms and software for operation, data treatment, and reporting. Following some years of uncertain future, during which parts of the facility became obsolete, JRC made the decision to revive the facility to support the ongoing nuclear D&WM programme. This decision coincided with the launch of MetroDECOM II project in 2017, giving the Nuclear Security Unit of JRC the possibility to participate by taking the two primary measurement stations of WCS into operation. Thus, it simultaneously provided support to the Decommissioning Unit of JRC and made the facility and results available to the wider European decommissioning and metrology community. Under MetroDECOM II, WP 3, the Nuclear Security Unit, together with their work package partner ENEA, took charge of the refurbishment of the two main measurement stations of WCS by implementing state of the art algorithms for data acquisition and analysis, conducting measurements on simulated waste packages for system validation, and demonstrating performance on real waste containers from the JRC-Ispra site.

The target, by the termination of MetroDECOM II in 2020, is to enable WCS to operate as the standard characterisation tool for radioactive waste on the JRC-Ispra site and as a demonstration facility in Nuclear D&WM for the European decommissioning and metrology community.

6.2 GAMMA AND NEUTRON STATIONS

The task of the Nuclear Security Unit and the MetroDECOM II project is limited to taking the two main stations into operation for 220 L drums only, and does not include the automation process, operation of the conveyor and drum movements.

The entire WCS system is designed to accommodate standard drums of 220 L and 400 L. A sketch of WCS, showing its principal components is shown in Figure 4 where the following five individual measurement stations for waste drums of WCS are visible:

- 1 Barcode reading/identification;
- 2 Weight measurement;
- 3 Dose measurement (at contact, and at one metre distance);
- 4 Segmented/tomographic gamma scanning (SGS/TGS);
- 5 Passive/active neutron measurement.

Not shown in Figure 4 is the entire building housing the WCS. The conveyer of WCS stretches almost the entire length of the building. Also not shown is the radiation shielded WCS control room adjacent to the WCS building which allows remote operation of all drum movements and measurement activities without staff present in the measurement hall.

The facility is designed for automatic operation of each measurement station and for movement of waste drums between stations. The on and off loading of up to 12 waste drums to the buffer zone on the conveyor is done by means of an overhead crane operated manually. In principle, the standard mode of operation could involve loading of 12 drums to be assayed in the afternoon, automatic and unattended assay of all drums in the five stations during night, unloading of the 12 drums in the morning of the following day. A cyclic procedure like this could see a throughput of 60 standard drums per week or 2000 drums per year.

The purpose of WCS is to deliver a comprehensive characterisation of each waste drum by producing assay results from the five stations presented in a single report and database entry on the basis of the following measurement steps:

1. The drum is identified by engraving a bar code on the exterior.
2. The weight of the drum is measured. The net weight of the waste is necessary for waste acceptance criteria established on waste activity concentration (quoted in Bq/g).
3. Values of dose measurements at contact, and 1 m distance, needed for transport regulations.

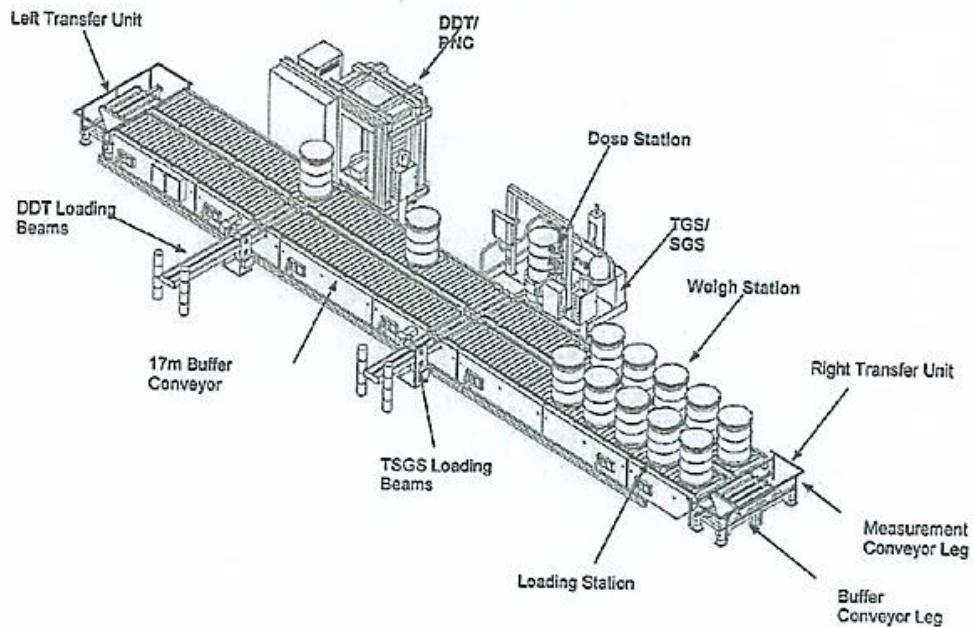


Figure 4 - Sketch of the WCS of the Nuclear Decommissioning Unit of JRC showing main components.

4. The gamma measurement station has the purpose to identify and quantify gamma emitting radionuclides in the waste by implementing both segmented gamma scanning (SGS) and tomographic gamma scanning (TGS), combined with transmission measurements using an external europium (^{152}Eu) source for estimating gamma-ray attenuation in the drum. In the standard SGS mode the drum is measured in 16 horizontal segments with and without the transmission source, while the drum is rotated. In the TGS mode each segment is measured in several voxels by means of both rotation and translation of the drum.
 5. The passive/active measurement station has the purpose to quantify the content of the two major actinides uranium and plutonium in the waste. This is done by passive neutron measurement techniques to observe spontaneous fission emitters, and an active measurement using an external pulsed neutron generator to induce fission in fissile isotopes with subsequent detection of the prompt fission neutrons. The combination of neutron measurements of certain fissile isotopes and the determination of the uranium and plutonium isotopic composition allows quantifying with good accuracy the content of uranium and plutonium in the waste item.
- The two major actinides are important alpha emitters thus their quantification is especially important for classifying the waste package according to (alpha) activity. Another key motivation for paying careful attention to these particular elements is their nuclear safeguards' concern. A robust method of characterisation is required for waste packages containing these elements for example in form of spent fuel residues or process waste from facilities concerned with uranium or MOX fuel fabrication in any of the many stages from enrichment of uranium or reprocessing of spent fuel to final fuel element fabrication. A recent IAEA guideline sets limits of masses and concentrations of uranium and plutonium in various kinds of waste for allowing the termination of safeguards inspections [3]. For facilities, producing this kind of nuclear waste, the guideline can be used to derive appropriate requirements to fissile mass assay of the WCS. Not surprisingly the limits set in the guideline suggest the need for substantial instrumentation in order to reach the levels for recommended termination of safeguards for the waste packages.

6.3 IT ARCHITECTURE

The WCS control and data analysis is performed by five interconnected computers:

1. FCC (Facility Control Computer);

2. MCC (Measurement Control Computer);
3. GSC (Gamma Station Computer);
4. WBBC (Weight Barcode Bridge Computer);
5. NSC (Neutron Station Computer).

A schematic design of IT architecture of WCS is reported in Figure 5.

All the computers, except the NSC, are installed in the control room of Waste Characterisation Facility (WCF) of JRC-Ispra site.

1. The FCC computer is directly connected to the Programmable Logic Controller (PLC) of the system. This computer shows the users a view of the entire facility status indicating if the system is in emergency stop status or not. The FCC computer is connected to the internal network of the system by an Ethernet switch in the control room. The user has the possibility to have direct control of all statuses of the different systems (FCC, MCC, GSC, WBBC and NSC) by remote visual systems (i.e. PC monitors).
2. The MCC computer allows performing the measurement in automatic mode. All the computers (WBBC, GSC and NSC) are internally connected by network switches. The MCC computer is also connected by the internal LAN to the measurement station. From this computer, the automatic mode measurements are performed at WCS. Besides, ANTECH installed the software for pre- and post-processing.
3. The GSC computer allows performing gamma and dose measurements. The computer is connected to gamma station and dose station by two switches: one in the control room and one in the measurement room. This computer is used to perform the gamma and dose measurements in manual mode, without using the MCC computer. On this computer are installed all the ANTECH 's software to perform gamma and dose measurements (*MasterScan*, *MasterAnalysis* and *MasterDose*).
4. The WBBC computer is only used remotely by a remote desktop application with the MCC. On the MCC is also installed the software necessary to perform the weight, bridge and barcode reader (*WeighServer*, *MasterBridge* and *MasterBarcode*). This computer is connected to the station by means of two switches, one in the control room and the other in the measurement room (see Figure 5).

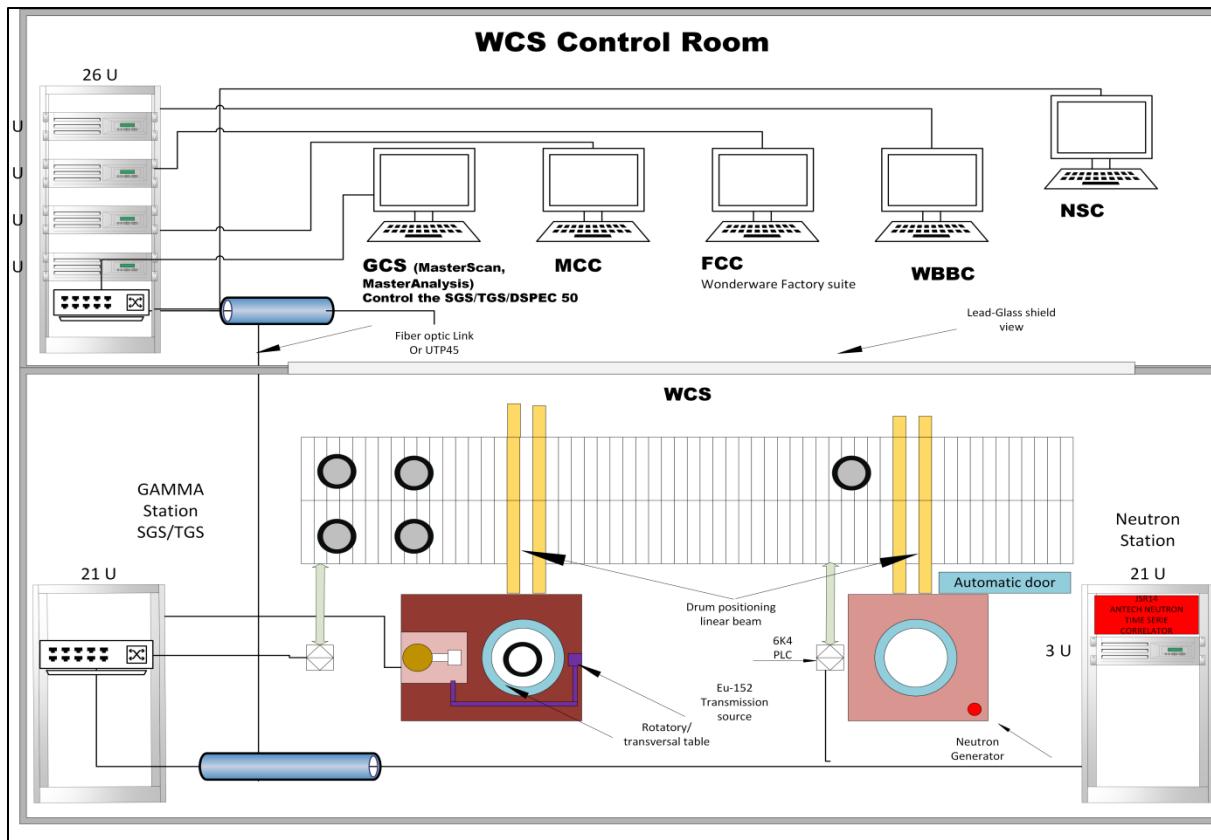


Figure 5 - IT architecture of the TSGS and NS systems at the WCS JRC-Ispra. The top is the control room, the bottom side the measurement room.

5. The NSC computer is installed close to the Neutron Station to minimize the distance from the electronics of the DDT generator as well the ${}^3\text{He}$ gas tubes.

7 METHOD VALIDATION ACCORDING TO ISO/IEC 17025:2017

7.1 ACCREDITATION OF TESTING LABORATORIES

Tests consist of the determination of the characteristics of a product in accordance with well-defined methods. All measurements carried out in the WCS system on waste drums can be regarded as tests and in this context the JRC-Ispra can be seen as a testing laboratory.

The ISO17025:2017 standard has been developed with the objective of promoting confidence in the operation of testing laboratories. The standard contains requirements for laboratories to enable them to demonstrate they operate competently, and are able to generate valid results.

In most Countries, ISO/IEC 17025 is the standard for which most laboratories can be deemed technically competent.

Many Countries have established "accreditation bodies" that can provide an external, third party, independent and formal recognition (accreditation) of competence that a testing laboratory perform tasks, i.e. to follow ISO/IEC 17025:2017 standard requirements in its operation. So, laboratories "accredited" under ISO/IEC 17025:2017 standard can demonstrate to their customers that they have been successful at meeting the requirements of international accreditation standards.

This is particularly important when tests are performed to show compliance with safety/regulatory standards on the basis of which the individual/environment protection is guaranteed.

Under this frame, the JRC-Ispra WCS activities are performed as far as possible, in compliance with the ISO/IEC 17025:2017 standard requirements.

7.2 ISO/IEC 17025:2017 REQUIREMENTS

The ISO17025:2017 standard contains different category requirements:

- General requirements;
- Structural requirements;
- Resource requirements;
- Process requirements;
- Management system requirements.

For the purpose of the present document, Resource and Process requirements are more relevant. Their structure is the following:

Resource requirements:

- Personnel (6.2);
- Facilities and environmental conditions (6.3);
- Equipment (6.4);
- Metrological Traceability (6.5).

Process requirements:

- Selection, verification and validation of methods (7.2);
- Selection and verification of methods (7.2.1);
- Validation of methods (7.2.2).

The present GPG will discuss most of the above Resource and Process aspects, except Personnel.

7.3 VALIDATION OF METHODS

7.3.1 Validation of methods

According to ISO/IEC 17025:2017 standard, the following definitions applies:

1. Verification: “Provision of objective evidence that a given item fulfils specified requirements”;
2. Validation: “Verification, where the specified requirements are adequate for an intended use”.

Therefore, validation is confirmation by examination and the provision of objective evidence that the particular requirements for a specific intended use are fulfilled. It is important to note that the specific intended use has to be specified and clearly stated when addressing any validation aspect.

In our context, the intended use can be regarded as the radionuclide or element to be measured, range of detectable and quantifiable total activity or mass, its distribution inside a drum, position and homogeneity, presence of any disturbing or interfering element, drum type and size, measuring time and so on. All these elements contribute to the definition of the scope of testing and the corresponding eventual scope of accreditation.

According to ISO/IEC 17025:2017 (7.2.1.3 NOTE) “International, regional or national standards or other recognized specifications that contain sufficient and concise information on how to perform laboratory activities do not need to be supplemented or rewritten as internal procedures if these standards are written in a way that they can be used by the operating personnel in a laboratory. It can be necessary to provide additional documentation for optional steps in the method or additional details”.

Unfortunately, in most of the cases, standard waste assay methods (§2.4) need to be adapted to the specific environment, scope of assay, equipment used, mathematical algorithms, waste characteristic. In all these cases, again, the following ISO/IEC 17025:2017 standard:

1. 7.2.2.1 "The laboratory shall validate non-standard methods, laboratory-developed methods and standard methods used outside their intended scope or otherwise modified. The validation shall be as extensive as is necessary to meet the needs of the given application or field of application".
2. 7.2.2.2 "When changes are made to a validated method, the influence of such changes shall be determined and where they are found to affect the original validation, a new method validation shall be performed".

Although measurement methods applied in the WCS system are described in several scientific papers and guides, it is not an explicit subject of any normative standard. Moreover, it is used with adaptations and modifications introduced by the system manufacturer as well as by the JRC researcher staff.

A validation (according to ISO/IEC 17025:2017 standard approach) is therefore required. The techniques used for method validation will be described in the following paragraphs.

7.3.2 Techniques for method validation

According to ISO/IEC 17025:2017 standard **7.2.2.1 NOTE2** "The techniques used for method validation can be one of, or a combination of, the following:

- b) calibration or evaluation of bias and precision using reference standards or reference materials;
- c) systematic assessment of the factors influencing the result;
- d) testing method robustness through variation of controlled parameters, such as incubator temperature, volume dispensed;
- e) comparison of results achieved with other validated methods;
- f) inter-laboratory comparisons;
- g) evaluation of measurement uncertainty of the results based on an understanding of the theoretical principles of the method and practical experience of the performance of the sampling or test method."

We have to note that validation not necessary has to use all the mentioned techniques. The tests performed at JRC on the WCS system will all belong to the above listed techniques, and although not exhaustive, will provide evidence for a system validation. Future work will look at application of the full list of the above listed techniques.

7.3.3 Performance characteristics

The Quality of a test can be quantitatively evaluated by its properties named: Performance characteristic.

According to ISO/IEC 17025:2017 standard:

- **7.2.2.3** "The performance characteristics of validated methods, as assessed for the intended use, shall be relevant to the customers' needs and consistent with specified requirements."
- **NOTE** Performance characteristics can include, but are not limited to,
 - a) measurement range,
 - b) accuracy,
 - c) measurement uncertainty of the results,
 - d) limit of detection,
 - e) limit of quantification,
 - f) selectivity of the method,
 - g) linearity,
 - h) repeatability or reproducibility,
 - i) robustness against external influences
 - j) cross-sensitivity against interference from the matrix of the sample or test object
 - k) bias.

Important to note that here too it is reconfirmed that performance characteristics (therefore the quality of the method) is evaluated with respect to the “intended use”, that therefore has to be clearly defined.

7.3.4 Records

A further important ISO/IEC 17025:2017 requirement regards the need to maintain records of the validation performed. This is used to provide objective evidence of the performed validation to external reviewers, accreditation assessors. Keeping well sorted records also helps to evaluate trends in system performance, improvements or decreasing quality, and as such represents an important complementary information.

According to ISO/IEC 17025:2017 standard:

7.2.2.4 “The laboratory shall retain the following records of validation:

- a) the validation procedure used;
- b) specification of the requirements;
- c) determination of the performance characteristics of the method;
- d) results obtained;
- e) a statement on the validity of the method, detailing its fitness for the intended use.

To some extent the present GPG represent a first record for the performed WCS validation. Following validation work can complete the present validation picture.

7.4 MetroDECOM II WP3 APPROACH TO VALIDATION

7.4.1 Approach

As described above, the aim of WP3 of MetroDECOM II, is to validate the new WCS developed at Ispra site for VLLW, LLW and ILW characterisation, regarding both the gamma (SGS and TGS) and neutron stations (Active and Passive).

The WCS, although not fully operative, was already available at JRC, so the validation process also involved, to some extent, testing of system operation.

For social, economic and scientific aspects, the extensive JRC Nuclear D&WM Programme, described under §4 above has to provide objective confidence in the JRC decommissioning operation. It is evident that this is exactly the same objective of the ISO/IEC 17025:2017 standard (“promoting confidence in the operation of laboratories”). Indeed, requirements for laboratories are set to demonstrate:

- competent operation;
- capability to generate valid results.

For this reason, the ISO17025:2017 approach to validation was followed as far as possible, in performing the validation work required in MetroDECOM II WP3.

7.4.2 Implementation

The WCS complexity and its multi component/method nature, in addition to the need to perform some initial system testing of operation, required more time than what was available in the MetroDECOM II project duration (3 years). In addition, a number of unexpected technical issues as well as the COVID-19 pandemic access to laboratory restrictions caused a considerable dilatation of the work programme.

For this reason, the validation work was not complete but the approach and examples given here represent an example of a Good Practice that is our intention to keep for the rest of the work.

The GPG will describe successful validation aspects as well as problems encountered including technical issues, as we consider these could be common to other laboratories (Table 5).

Table 5 - Summary of validation methods and performance characteristics applied or determined in the present work

| | SGS | TGS | Passive Neutron station |
|---|--------|-------|-------------------------|
| VALIDATION METHODS | | | |
| a) calibration or evaluation of bias and precision using reference standards or reference materials | 9.3.4 | | |
| b) systematic assessment of the factors influencing the result | 9.3.7 | | 12.2 |
| c) testing method robustness through variation of controlled parameters | | | |
| d) comparison of results achieved with other validated methods | 9.5 | | |
| e) inter-laboratory comparisons | | | |
| f) evaluation of measurement uncertainty of the results | | | |
| PERFORMANCE CHARACTERISTICS | | | |
| a) measurement range | 9.3.1 | 9.3.2 | 8.2.2 |
| b) accuracy | | | 11.3.9 |
| c) measurement uncertainty of the results | 9.5 | 10.1 | 11.3.9 |
| d) limit of detection | 9.3.12 | | 11.4 |
| e) limit of quantification | 9.3.12 | | 11.4 |
| f) selectivity of the method | | | 11.3.1 |
| g) linearity | 9.3.1 | 9.3.2 | 11.3.10 |
| h) repeatability or reproducibility | | 9.3.8 | 11.4 |
| i) robustness against external influences | | | 11.3.7 |
| j) cross-sensitivity against interference from the matrix of the sample or test object | | | 11.3.4 |
| k) bias | 9.3.13 | | 11.4 |

7.4.3 The GPG and the view for future accreditation

During the execution of the MetroDECOM II project, the JRC Ipra laboratory developed a good experience oriented to ISO/IEC 17025:2017 management. This will be useful in the future for the JRC Nuclear D&WM Programme and for other laboratories training. This experience is described in the present GPG.

The ISO/IEC 17025:2017 approach to validation will be hopefully extended to other aspects of the JRC laboratory operation with the aim of reaching formal accreditation in the near future.

8 REFERENCE MATERIALS

8.1 SIMULATED WASTE DRUMS

An investigation was initially carried out on the types of waste matrices currently present on the JRC-Ispra decommissioning site. They are packed in 220 L cylindrical waste container (drum) which has become the standard waste container on the site [10].

In order to carry out the validation (including calibration) and Quality Control work, a number of simulated waste drums were developed that represent and reflect, as far as possible, the various matrix types encountered in the real wastes at JRC. The simulated waste containers can be filled with various non-active matrix material and have re-entrant tubes for the

insertion of reference standard sources or Reference Materials at well-defined locations within non-active material.

Five simulated waste drums, were thus prepared by the Ispra Waste Management Support (IWMS) Consortium [11] plus, one empty drum for the purpose of system calibration and characterization, as reported in Table 6 and shown in Figure 6.

Table 6 - Simulated waste drums prepared for WCS validation.

| No. | Abbreviation | Matrix material | Density range (g/cm3) | Filling material | Mass (kg) | Label drum and spacers |
|-----|--------------|-------------------------------------|-----------------------|--------------------------------|-----------|------------------------|
| 0 | EMT | Air | - | Air | 51.8 | Black |
| 1 | TCO | Technological Waste combustible | 0.7 - 1.0 | Paper, Plastic, cotton, Tyvek® | 91.4 | Red |
| 2 | TNC | Technological Waste Non-Combustible | 2.2 - 2.4 | Glass | 170.8 | Yellow |
| 3 | CMT | Cemented waste | 2.7 - 3.0 | Portland cement | 468.8 | White |
| 4 | RBL | Rubbles waste | 2.5 - 2.8 | Rubbles, graves, stones | 261.4 | Light Blue |
| 5 | MTL | Metallic waste | 6.0 - 8.0 | Mixed metal (no lead) | 253.0 | Pink |

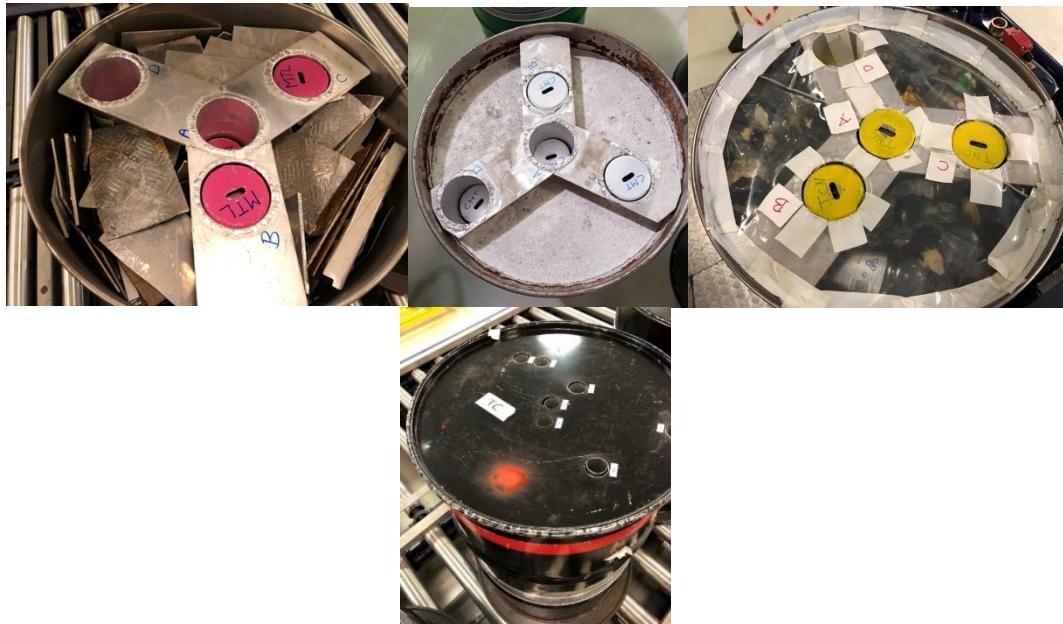
The five calibration drums were produced in order to be as homogeneous as possible. Each calibration drum was filled with the corresponding material and the mass of the calibration drum is given combining the spaces (i.e. channels and hollow container in Figure 6) and the drum itself. Each measured drum is identified by a barcode corresponding to the internal reference number, so called WITS number.



Configuration 1 used for the gamma station.



Configuration 2 used for the neutron station.



Simulated drums in clockwise direction from top left: MTL, CMT, TNC, and TCO.

Figure 6 - (On the first row) Standard 220 L drum for TSGS characterisation with 7 entrance channels, indicated as the “empty” drum. **(On the second row)** Reference empty 220 L drum with aluminium carrot holder of different sides for Pu CBNM standard sources in aluminium structure with 4 entrance channels (on the left picture). The ‘carrot’, a hollow aluminium cylinder, is able to position the CBNM standards at three different height (on the right picture). **(On the third row)** The simulated drums for the used TSGS characterisation.

8.2 STANDARD SOURCES AND CERTIFIED REFERENCE MATERIALS

8.2.1 Standard sources for gamma station

A total of 12 single radionuclide standard point sources, with calibration certificates, were used for WCS validation. Sources were encapsulated in aluminium enclosure for safe operation and prevention of radioactive contamination (Figure 7).



Figure 7 - Picture of the Cs-137 and ^{60}Co single radionuclide standard point sources.

Source radionuclides were ^{241}Am , ^{133}Ba , ^{137}Cs , ^{60}Co and ^{152}Eu to cover and define the system operating photon energy range. Calibration certificates specify values of each radionuclide activity at a given reference date, as reported in Table 7.

The selected radionuclides also represent pre-selected nuclear vectors identified by the TSGS.

Relevant data of these sources were entered in the pre or user defined Calibration Standards screen accessible from the main menu in *MasterScan* program. If a calibration standard is not defined it is possible to create a new one through the *MasterScan* menu.

Table 7 - Standard radioactive sources used for validation of the gamma station of the JRC-Ispra WCS.

| Source ID | Radionuclide | Activity (Bq) |
|-----------|-------------------|---------------|
| 579 | | 1.74E07 |
| 181 | ^{241}Am | 3.70E05 |
| 97 | | 4.14E05 |
| 613 | | 3.89E05 |
| 95 | ^{133}Ba | 3.27E05 |
| 580 | | 5.00E05 |
| 719 | ^{137}Cs | 9.17E05 |
| 307 | | 3.65E05 |
| 212 | ^{60}Co | 3.70E07 |
| 310 | | 3.70E05 |
| 544 | ^{152}Eu | 2.29E06 |
| 385 | | 1.85E08 |

8.2.2 Certified Reference Materials for Neutron station

The Pu sources used in the neutron measurement validation campaign are nuclear Certified Reference Materials. The source material is PuO_2 and they were certified on 20th June 1986 by the European Central Bureau for Nuclear Measurements (CBNM). The characteristics of the samples are given in

Table 8, displaying for each sample the plutonium mass, the Pu isotopic composition and ^{241}Am content updated to date of measurement campaign. The table also lists the total Pu mass, and the ^{240}Pu effective, $^{240}\text{Pu}_{\text{eff}}$, mass calculated according the following formula:

$$\text{Mass}(\text{Pu}_{\text{eff}}) = \text{Mass}(Pu) \times (2.66f^{238} + f^{240} + 1.64f^{242}), \quad (1)$$

where the f^m are the weight fraction of the plutonium isotope with atomic mass number m.

Table 8 - CBNM PuO₂ Certified Reference Materials used in the passive neutron measurement validation campaign.

| Sample code | Pu mass [g] | ²⁴⁰ Pu _{eff} mass [g] | Alpha ratio |
|-------------|-------------|---|-------------|
| Sample 1 | 5.820 | 0.372 | 0.814 |
| Sample 2 | 5.830 | 0.877 | 0.568 |
| Sample 3 | 11.650 | 1.249 | 0.641 |
| Sample 4 | 5.536 | 1.357 | 1.193 |
| Sample 5 | 5.426 | 1.997 | 1.013 |
| Sample 6 | 11.365 | 2.233 | 0.948 |
| Sample 7 | 10.961 | 3.354 | 1.086 |

9 VALIDATION OF GAMMA STATION TSGS

9.1 SYSTEM CHARACTERISTICS

9.1.1 System description

The gamma measurement station has the function, when the drums are loaded into the measurement position, to measure the surface dose rate and drum content of gamma emitting radionuclides, in one of both two analysis modes: SGS or TGS. In the report the acronym TSGS (Tomography Segmented Gamma Scanning) will be used to refer to both scanning modes SGS/TGS, without need to differentiate.

The operator is able to choose the scanning mode through the MCC that is linked to the individual measurement instrument computers.

The surface dose rate is measured by 6 Geiger Muller (GM) counters positioned around the drum. Three of them measure the dose on the side, one the dose above and one the dose below the drum. The last GM counter is positioned to measure the dose at a distance of 1 m from the drum surface.



Figure 8 - Pictorial view of the gamma measurement station from different points of view: from the beam loader (on the left), from the acquisition electronic chain side (on the right) (Source: JRC, 2019).

The principle TSGS components are (Figure 8):

- 1 plinth with levelling and fixing points;
- 2 pillar with vertical scan slide that supports the HPGe detector, the liquid nitrogen dewar and an arm that holds the ^{152}Eu transmission source;
- 3 liquid nitrogen dewar;
- 4 rotating table on horizontal slides that receives the drum, takes it to its measuring positions, for dose rate and TSGS measurements, and provides the horizontal and rotation motions for the scan process;
- 5 shielded ^{152}Eu source support;
- 6 ^{152}Eu , 1 GBq, transmission source holder with shutter;
- 7 drum support to allow drum loading with beams (220 L / 400 L drums resting on pucks);
- 8 facility electric interface wiring cubicle;
- 9 control cabinet housing motor drives etc.

Those components are the same used either for the SGS or for the TGS mode.

9.1.2 System characteristics

At the beginning of the loading operation, the operator has to set the measurement mode and choose the correct type of tungsten collimator to put in front of the detector: square or diamond shape. The square shape is used for the SGS mode while diamond shape for the TGS mode (i.e. one for a 220 L and another one for 400L drum). Both tungsten collimators are inserted in a lead shielding around the Ge crystal.

The electronic Gamma-ray chain consists of the following:

- 1 ORTEC GEM series High-Purity germanium (HPGe) coaxial detector with a crystal size of 65.8 x 72.3 mm, 50% relative efficiency and FWHM of 0.85 keV (at 122 keV ^{57}Co gamma line);
- 2 ORTEC DSPEC50® Digital Signal Processing Gamma Spectrometer with Ethernet/USB connectivity to the PC Master Scan system (16k ADC Conversion Gain, List mode, single MCA and high count rate applications capabilities);
- 3 ORTEC 419 Precision Pulser Generator that generates a 1974.5 keV equivalent electronic pulse for energy stabilization and dead time correction;
- 4 ORTEC MAESTRO acquisition system;
- 5 Optical Ethernet connectivity to the FCC.

9.1.3 Analysis methods SGS

In the case of SGS mode two conditions must be met to optimize assay results as follows [7]:

1. The particles containing the nuclides of interest must be small to minimize self-absorption of emitted gamma radiation.
2. The mixture of material within a package segment must be uniform in order to apply an attenuation correction factor, computed from a single measurement of item transmission through the segment.

For simplicity let us assume a 220 L waste drum that contain one single radionuclide X_i . The following peaks will be observed on the gamma spectra:

- Pulser¹ peak (emission and transmission);
- Radionuclide X_i emission peak²;
- ^{152}Eu transmission peak.

For each of these peaks, a ROI is identified with the following quantities calculated:

- NP : Number of channels of the ROI;

¹ Pulser indicates the digital signal pulse inserted in the electronic chain for dead-time correction.

² Emission peak of a radionuclides X_i is referred always as the FEP (Full Energy Peak).

- P : Peak counts (sum of counts in the ROI);
- NB_1 : Number of Background channels on the left of the ROI;
- NB_2 : Number of Background channels on the right of the ROI;
- B_1 : Background counts on the left of the Peak;
- B_2 : Background counts on the right of the Peak.

For each scan layer the following peaks (identified with letters a) to g)) are recorded and analysed:

| | Phase | | | |
|-------------------|----------|--------------|------------------|------------------|
| | Emission | Transmission | Straight-through | Straight-through |
| Pulser | (a) | (c) | gh | through |
| Radionuclide | (b) | | shutt | shutter |
| X_i | | | er | closed |
| ^{152}Eu | | (d) | open | |
| | | | (e) | (g) |
| | | | (f) | |

The Net Peak Area, A , for all mentioned peaks (radioisotope X_i , pulser and ^{152}Eu transmission source) is calculated by the formula:

$$A = P - \left[\frac{NP}{2} \right] \left(\frac{B_1}{NB_1} + \frac{B_2}{NB_2} \right). \quad (2)$$

with:

- T_s : counting time of the Straight-through measurements;
- T_g : counting time of the grab measurements (emission/transmission);

The Net Peak Area, C_{ST} , for all mentioned peaks, normalized to the same grab measurement time, is given by:

$$C_{ST} = A \times \frac{T_s}{T_g}. \quad (3)$$

with:

- $L T_0$: pulser peak count rate in Straight-through condition (g) above;
- $L T_i$: pulser peak count rate in other conditions (a) to f)) above.

The Net Peak Area, A'_i , corrected for dead time losses, is given by:

$$A'_i = C_{ST} \times \frac{L T_0}{L T_i}. \quad (4)$$

Photon attenuation in the container, for each i-th layer, is matematically described by the Container Transmission Single Correction Factor, T_c , defined as:

$$T_c = \left(\frac{TA'_i}{TA'_0} \right)^{KA}, \quad (5)$$

where:

- TA'_i : corrected Net Peak Area (Eq. 2 to 4) of the transmission with empty container (no matrix in the drum) for each i -th layer;
- TA'_0 : corrected Net Peak Area (Eq. 2 to 4) of the Straight-through transmission source with no container;
- KA : linear attenuation coefficient for the specific container type and photon energy.

The TA'_i middle values (8-layer) for each gamma peak of the ^{152}Eu transmission source are stored in the database. The user will need to enter the linear attenuation value for the container type and mass attenuation value for the material type at each specific energy line. Thus the T_c values will be calculated for each layer and energy line of interest by Eq. 5. The Percent of Straight-through is also stored in order to be used to check for dense segments.

Two conditions are also stored:

1. if the Transmission Corrected Net Peak Area, $A'_i < \text{Percent of Straight-through}$ for the same Transmission No container Straight-trough Corrected Net Peal Area then we A'_i and to 1E300 ("Density Percent" error is noted).
2. if $A'_i < 0$, or $TA'_0 < 0$ for a particular transmission energy line then a "Density Zero" error is noted by the program. Then the transmission energy line is not used in the calculation and the calculation move to the next closest transmission energy line for the evaluation.

Then we calculated the Contents Transmission Single Correction Factor, T'_i , using the KA and T_c from (5):

$$T'_i = \frac{\left(\frac{TA'_S}{TA'_0} \right)^{KA}}{T_c}, \quad (6)$$

where:

- TA'_S : corrected Net Peak Area (Eq. 2 to 4) of the transmission with container filled with a matrix, for each i -th layer. (From Eq. 5), the Contents Attenuation factor, $CF_i(T'_i)$, and the Container Attenuation factor, CF_{can} , are derived:

$$CF_i(T'_i) = \frac{-B \times \ln T'_i}{1 - (T'_i)^B}, \quad (7)$$

and

$$CF_{can} = \frac{1}{\sqrt[2]{T_c}}, \quad (8)$$

where:

- B : geometry factor (0.823 for cylinder) x Mass Attenuation at energy line i .

Combining equations above we finally derive the Corrected Count Rate, CC_i , or SGS Number (SGS#), for each layer i :

$$CC_i = \frac{A'_i}{t} \times CF_i(T'_i) \times CF_{can}, \quad (9)$$

where t is the counting time.

The variance of the Net Peak Area $\sigma^2(A)$ is given by:

$$\sigma^2(A) = P + \left[\left(\frac{NP}{2 \times NB_1} \right)^2 \times B_1 \right] + \left[\left(\frac{NP}{2 \times NB_2} \right)^2 \times B_2 \right], \quad (10)$$

with all quantities already defined.

The $\sigma^2(A)$ needs to be corrected to the Straight-through time length, as in Eq. (2), and calculate the relative variances, $\sigma_r^2(A)$, for the emission and transmission records (see points 1 and 2 on list page 31):

$$\sigma_r^2(A) = \left(\sigma_c^2(A) / A \right)^2, \quad (11)$$

where $\sigma_c^2(A)$ is the corrected variance from Eq. 9).

The relative variances are then obtained:

- $\sigma_r^2(L_0)$: for the emission record pulser peak;
- $\sigma_r^2(A_0)$: for the emission record peak;
- $\sigma_r^2(T_0)$: for the transmission record (see list on page 31).

The system also defines a K factor as:

$$K = \left(\frac{B}{CFAT} \right) \times \left[\frac{\left(1 - (TR^B \times CFAT) \right)}{1 - TR^B} \right], \quad (12)$$

where:

- $CFAT = CF_i(T_i)$;
- $TR = (T_i / T_0) \times (CF_{can}^2)$;

used to calculate the Corrected Count Rate Variance, $\sigma^2(CC_i)$, for the 3 gamma energy peaks:

$$\sigma^2(CC_i) = CC_i^2 [\sigma_r^2(A_0) + K^2 \times \sigma_r^2(T_0) + (1 - K)^2 \times \sigma_r^2(L_0)], \quad (13)$$

Thus $\sigma(CC_i)$ divided by Live Time is the variance error for the SGS number (SGS# number $\pm \sigma(CC_i)$). Then final SGS number is the sum of all layer contributions (i.e. 16 layers), ($SGS\# = \sum_i CC_i$ from eq. (8)). The activity of the container is the product of the SGS number and the Calibration Factor:

$$Activity(X_i) = SGS\# \times CF \quad (14)$$

9.1.4 Calculations of detection limit for the TSGS measurement

The SGS system evaluates for each layer i a detection threshold, DT so defined:

$$DT = \frac{1}{\sqrt{\sum_i \frac{1}{\left(\frac{X}{T} \times CF_i(T'_i) \times CF_{can} \right)^2}}}, \quad (15)$$

where the all quantities in the eq. (15) has already been defined above and the X is determined by this formula:

$$X = 1.96 \sqrt{(B_1 + B_2) \times \left(\frac{NB_1 + NB_2}{P} + 1 \right)}, \quad (16)$$

with B_1 , B_2 , NB_1 , and NB_2 above. Only if the absolute value of the SGS# is greater than DT that the detection limit (DL) is determined as:

$$DL = 2 \times DT \quad (17)$$

9.2 Analysis method TGS

TGS delivers a better performance over SGS for the characterisation of radioactive waste drums. In SGS, we assume a uniform attenuation coefficient for each layer. TGS allows to measure the attenuation map for each layer and then to correct for self-attenuation.

The process comprises two parts:

- 1 determine the attenuation map resulting from the measurement of the ^{152}Eu source (transmission);
- 2 determine the matrix emission map from the internal radionuclides (emission).

Many algorithms are developed to reconstruct the image from the projections ³. Unfortunately, in TGS, the number of counts per measurement point (and for each energy line) is too low during a waste drum standard measurement (i.e less than 8 h).

To face this problem, ANTECH has implemented a statistical approach which gives better performance. The traditional ART method is also used in TGS and has been implemented in the TGS_FIT program.

9.2.1 Reconstruction problem

In order to reconstruct an image of the drum matrix and its content (i.e. hot spot) we consider that the drum is divided in n layers (e.g. 16). For each layer, and point at coordinates x , y , z , the attenuation factor $\mu = \mu(x, y, z, E_i)$ must be determined, where E_i is the emission energy of the radionuclide of interest i . The attenuation map, μ , is reconstructed from the projections resulting from the rotation and translation of the detector, relative to the drum (as showed for the gamma measurement station, see Figure 9).

³ Filtered Back Projection (FBP), Algebraic Reconstruction Technique (ART), Simultaneous Iterations Reconstruction Technique (SIRT) algorithms of the ART family.

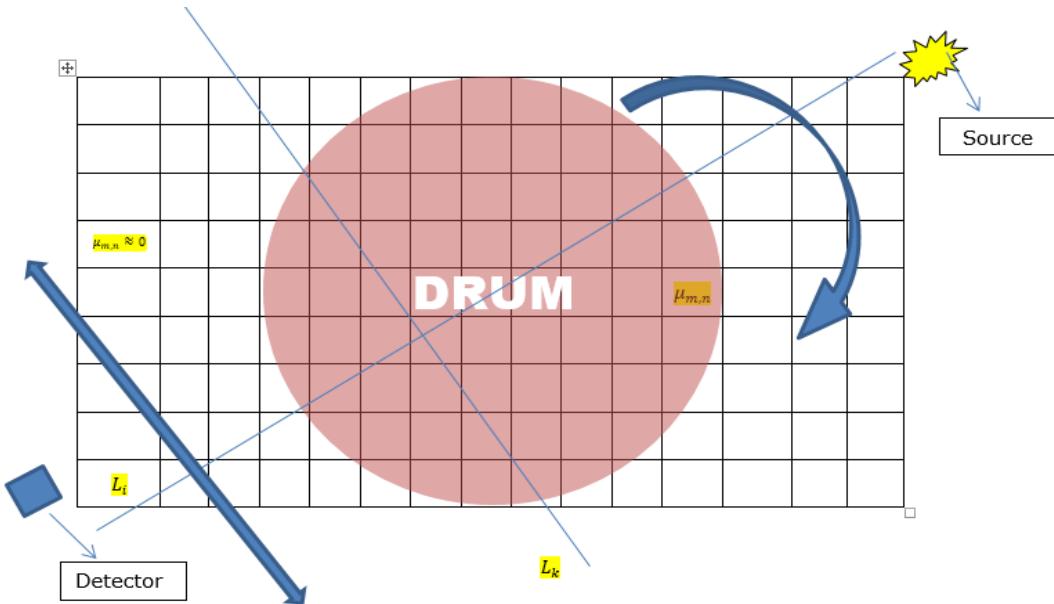


Figure 9 - Schematic representation of a waste drum (red circle) under the tomography scan (the grid) with rotation/translation movement between the line from the HPGe detector (bottom left corner) and the transmission source (top right corner). L_i and L_k are the projection line over which the μ is summed (Radon transform).

This process will generate N_y measurements, where N_y is the product of the number of translation times the number of rotations. For each measurement point (and for each E_i), the average number of photons recorded by the HPGe detector is ideally expressed through the Beer–Lambert law⁴, and we can derive the intensity or number of photons, I :

$$I = I_0 \times \exp(-\int \mu(x, y, E_i) dl), \quad (18)$$

where I_0 is the count rate for a point source at a given energy and the attenuation map is integrated along L_i , the line path length through the material sample (matrix) (Figure 9). We denote by b_i the l value measured by the detector when the matrix is empty, or $\mu = 0$, and the known source is in position (x, y) . b_i is equivalent to I_0 value or activity of the source but corrected for the geometry, solid angle, detector efficiency, etc. For example, if the activity of the source is 100 Bq (with photon emission intensity and counting efficiency equal to 1) and we have a detector surface of 1 cm^2 placed at 1 m from the source, the detector will measure $100/(4\pi \times 10^4)$ counts/s.

Let us call $Y_{i,}$ a measurement value, that is the random variable representing the detector count rate for the i -th photon. Then from eq.(2) we can express the number of counts for a specific energy (E is the expected value of Y_i) by:

$$E[Y_i] = I_0 \times \exp(-\int \mu dl) + r_i, \quad (19)$$

where r_i is the background count rate for the event number i . Y_i and r_i follow a Poisson distribution. For each layer and for each energy line, we will have N_y measurements points.

This set of N_y measurement is called a sinogram: $\{Y_i = y_i\}_{i=1}^{N_y}$.

We assume that the real value $l_i^{true} = \int \mu dl$. Applying logarithim function on both terms of eq.(18), the measurement Y_i provides an estimation of the line integral, \hat{l}_i , on i -th ray (with $I_0 = b_i$):

⁴Beer–Lambert law relates the attenuation of light to the properties of the material through which the light is travelling.

$$\hat{l}_i = \begin{cases} \ln\left(\frac{b_i}{E[Y_i] - r_i}\right), & Y_i > r_i, \\ \text{unknow}, & Y_i \leq r_i \end{cases} \quad (20)$$

so that we can determine $E[\hat{l}_i]$. We have to note that $E[\hat{l}_i] > l_i^{\text{true}}$ based on Jensens inequality⁵. This shows that the observed \hat{l}_i systematically over-estimates the true line integral value [12].

The grid above shows the attenuation map where each cell C_{ij} is characterised by a constant attenuation factor μ_{ij} (for each energy line).

This attenuation map can be parametrized in a discrete way (from eq.2) in the following way:

$$\vec{\mu} = \sum_{j=1}^{N_p} \mu_j X_j(x, y), \quad (21)$$

where N_p is the number of voxels in the map (like a grid of 10×10 cell $n \times m$), $X_j(x, y)$ is a basis function⁶ defined as a rectangle function $\text{rect}(t)$:

$$\text{rect}\left(\frac{x-x_j}{\Delta}\right) * \text{rect}\left(\frac{y-y_j}{\Delta}\right), \text{ with } \text{rect}(t) = \Pi(t) = \begin{cases} 0 & \text{if } |t| > 1/2 \\ \frac{1}{2} & \text{if } |t| = 1/2, \\ 1 & \text{if } |t| < 1/2 \end{cases} \quad (21)$$

where Δ is the size of the voxel (a two dimensional size).

If we reshape the attenuation matrix (Eq. above) into a vector, $\mu_{kl} \rightarrow \mu_j$, we can write the attenuation map $\underline{\mu} = [\mu_1, \dots, \mu_{N_p}]$, that is linked to the sinogram: $\underline{Y} = [Y_1, \dots, Y_{N_Y}]$.

Using the eq. $E[Y_i] = I_0 \times \exp(-\int \mu dl) + r_i$, (19) we can write:

$$l_i^{\text{true}} = \int \mu dl = \int \sum_{j=1}^{N_p} X_j(x, y) dl = \sum_{j=1}^{N_p} \int X_j(x, y) dl = \sum_{j=1}^{N_p} a_{ij} \mu_j, \quad (22)$$

or simplify in matrices product, where $A = \begin{pmatrix} a_{11} & \cdots & a_{1i} \\ \vdots & \ddots & \vdots \\ a_{j1} & \cdots & a_{ji} \end{pmatrix}$

$$A \times \vec{\mu}. \quad (23)$$

A is a sparse matrix⁷ (usually very large) and it is called projection matrix. In summary, to solve our l_i^{true} value we will have to produce a matrix from the emission reconstruction (A) and one vector from the transmission information ($\vec{\mu}$) (i.e. attenuation map).

In order to obtain the emission reconstruction A we have to determine an emission map from the recorded counts at the HGPe detector for all energy lines of interest.

In the following section we will show the connection between this mathematical formulation and the ANTECH softwares at the WCS.

9.2.2 WIN_TGS/TGS_FIT/TGS_VIEW code

⁵ Jensens inequality relates the value of a convex function of an integral to the integral of the convex function. The inequality states that the convex transformation of a mean is less than or equal to the mean applied after convex transformation.

⁶ It is an element of a particular basis for a function space.

⁷ The matrix A can be corrected to account for the length of the segment of the line L_i crossing the cell C_{kl} .

In the WCS the gamma measurement station has a software suite installed by ANTECH to perform various tasks. In particular for the TGS (in a more limited way also for SGS) the system uses the WIN_TGS, TGS_FIT and TGS_VIEW software suite written by R. Estep [13]. Mainly the reconstruction is performed by TGS_FIT software the others on the suite provides GUI interface to help the user with the setting of the drum tomography's reconstruction parameters. TGS_FIT is complex and difficult to use, and requires either very detailed instructions or a good background in physics of TGS measurements to be used. For this reason to simplify the commands and instruction for a facility operator, the reconstruction code has been interfaced with a text-based interface by the WIN_TGS (i.e. Microsoft Windows® application).

As mathematically derived in Eq. 21, the most important matrices are the transmission matrix (T -matrix), the emission matrix set (E -matrices), and the attenuation-correction thickness table (A -matrix). The reconstruction of the base Emission and Transmission for the set of scan geometries (e.g. number of views per layer, voxels per layer, and layers per sample) is performed by the MAKE_MAT program and then it feeds them into the TGS_FIT program. All this is performed through a batch type window program (e.g. *Analyze.bat*) that will call separate routines to have all the setting parameters for TGS_MAT and be finally able to provide E - and T - matrices. Following is an example of list of file (settings file for the specific TGS measurement) used by TGS_MAT:

- DSPECPLUS246+Am.lis
- TGS_220_16.set
- TGS_220_16.sgp
- DSPECPLUS246+Am.zmt

The TGS_MAT software (Figure 10) will let the user to define many parameters (e.g Scanner, Collimation and Offset) on the TGS measurement that will be very important and essentials to be used as inputs (i.e. file SGP) for the TGS_FIT program.

The TGS_FIT performs different analysis that they can be summarized with the following:

- The t-data (T -matrix) is converted to u-images (one layer at a time) using non-negative least square (NNLS) algorithm;
- The u-images are combined with the base E-matrices to build the full drum, attenuation-corrected emission matrix, F;
- Target-layer partitions of the F-matrix are applied to e-data to generate initial values for the s-images, using NNLS algorithm (i.e. single layer fits are performed);
- Using the full F-matrix with the s-vector pre-images as a starting point, the full drum emission image is reconstructed using either the Algebraic reconstruction technique (ART) or the Expectation Maximization (EM) algorithm.

The full drum image reconstruction serves to consolidate the source images into their correct layers by explicitly treating cross-layer emission.

TGS_FIT features three images-reconstruction routines:

- a) NNLS, a constrained fitting function for creating transmission images (u-images) and for generating single-layer emission preimages that are used at initial values for the full-drum emission fit;

For the full drum emission reconstruction two methods are used:

- b) Algebraic reconstruction technique (ART) [14]
Expectation maximization (EM) [15]

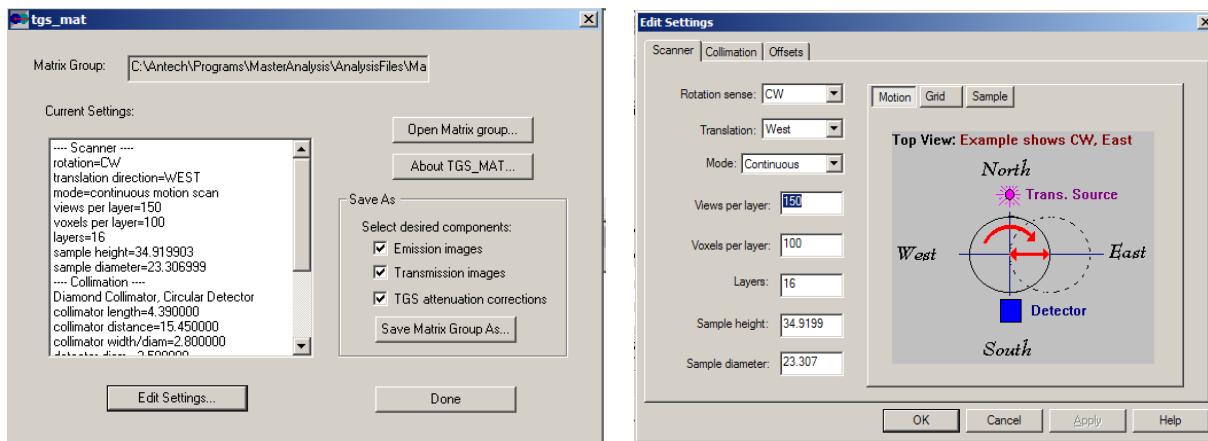


Figure 10 - TGS_MAT software with the Edit Settings on the right side for a TGS.
(Source: JRC, 2019).

It is important to notice that the current version of the TGS_FIT include recent developments on the material basis set (MBS) model and the gross count MBS (GC-MBS). The recent GC-MBS (or GC-TGS) method uses all counts in the spectrum above approx. 100 keV to produce an emission image and radionuclides mass with little sacrifice in accuracy [13]. It is understood that this will produce a limitation on analysis for radionuclides that have an energy <100 keV.

9.3 METHOD VALIDATION

According to ISO/IEC 17025:2017 standard (see §7.3.2) and in the method validation context, to characterize the TSGS, a set of measurements on simulated waste (e.g. calibration drum) has been performed. They addressed the following:

1. radionuclide to be measured;
2. range of detectable and quantifiable total activity;
3. its distribution inside a drum;
4. position and homogeneity;
5. presence of any disturbing or interfering element;
6. drum density;
7. measuring time;
8. repeatability of the measurement and so on.

SGS requires an initialization work before to get the system ready to receive a drum for a scan. This consists in an efficiency calibration of the gamma detector and in a determination of the waste matrix density.

Usually, these calibration works are required for all types of matrix and all radionuclides although in some cases some calculations can be used instead (i.e. MCNP) to allow self-absorption correction.

Efficiency calibration is performed with dedicated drums containing various density wastes in which known radioactive sources are inserted at various positions through vertical channels in the drum.

9.3.1 INIT for SGS

In order to perform measurements to calculate the activities of unknown radioactive sources the SGS system has to be initialized (i.e. INIT) with a calibration procedure.

The INIT was performed with a 400 ms measurement in an empty drum at the central vertical axis of the rotatory table (the middle centre of a drum sample, aka position A3), about 42 cm from the bottom of the puck. The calibration was performed using four radionuclides: ^{241}Am , ^{133}Ba , ^{137}Cs , and ^{60}Co (Figure 11). These cover the energy range for most of the common

radionuclides in nuclear facility waste. Radioactive sources were put on four different channels (A, C, G, and I, Figure 6) of the standard waste drum to simulate the assumption of uniformity of the absorption coefficient in the drum matrix (a set of 6-7 linear radioactive sources should be used for precise calibration instead, for availability purpose, we used point sources to calibrate the reference matrix's drums).

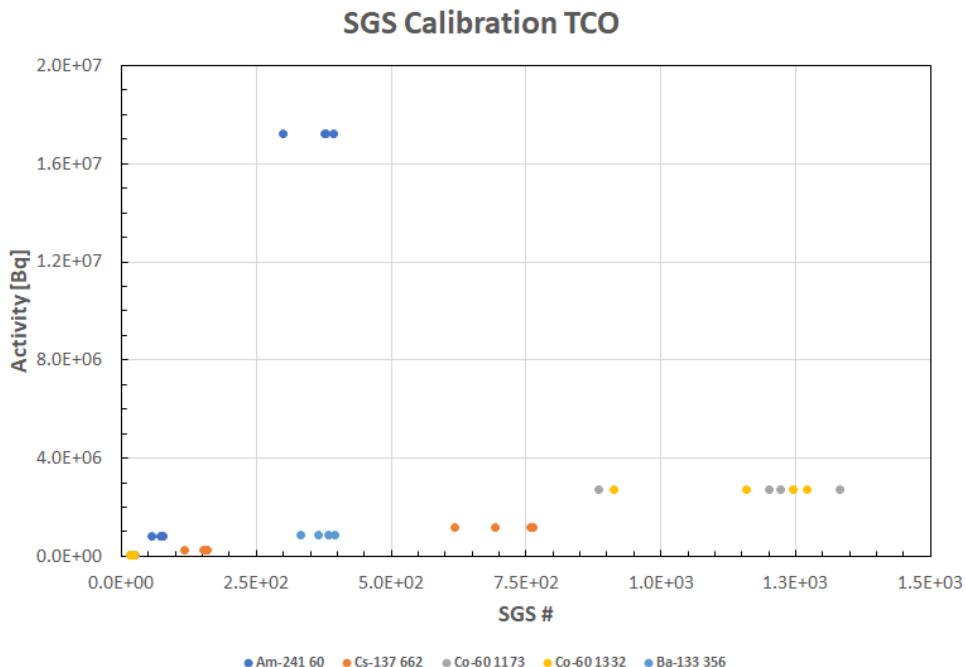


Figure 11 - Example of a calibration curve for the SGS mode for a TCO matrix with four radionuclides and their energy of interest.

9.3.2 INIT for the TSGS

In the case of the TGS mode a single calibration based on a non-interfering or empty matrix is made for several radioactive sources and different range of activities in order to cover the largest calibration spectrum of the real drums. The TGS system will use those calibration curves (i.e. “mass calibration”) in order to correct the attenuation information, determined from the transmission scans during a TGS measurement.

In this section we will describe the SOP to actually perform a TGS acquisition scan for calibration purpose at gamma measurement station of WCS. This procedure also contains the information to perform the analysis of the raw data once the TGS calibration scan is completed (determination of the activity of the radionuclides of interest and visualization of density matrix with the hot-spot position). All of this is performed using the Gamma measurement Station Computer (GSC).

In order to prepare the system for TGS measurement a calibration SOP needs to be followed:

1. Put on the gamma measurement station of WCS an empty drum containing only a chosen standard radionuclide source in its centre. The calibration has to be performed with one source at a time.
2. Check on Maestro® for Windows program the right operation of the HPGe detector chain.
3. Open MasterScan program from Start, if it is not already running.
4. In MasterScan menu select SETUP and SCAN PARAMETER sub-menus and set the parameters of the case (Figure 12):
 - a) Define the Grab Time in units of ms. The grab time defines the total desired scan acquisition time (i.e. with a 3000 ms as grab time one will have a 4 h total scan);

- b) Click on Calibration tick and select the desired standard source on the list available in Calibration Standard. Most of the common radionuclides are presented with the available radionuclides in the storage;
 - c) Select Emission/Transmission on Scan Type and TGS on Measurement Type.
 - d) Click on AUTHORIZE SCAN.
5. Click on Run in MasterScan menu and on the new window (Figure 12):
- a) Write a name for the measurement in Sample ID;
 - b) Select the Assay Type to the ones available on the list (i.e. TGS_220L_16_13082019);
 - c) Write the weight of the drum +22kg of the drum holder on Gross Sample Weight;
 - d) Click START.

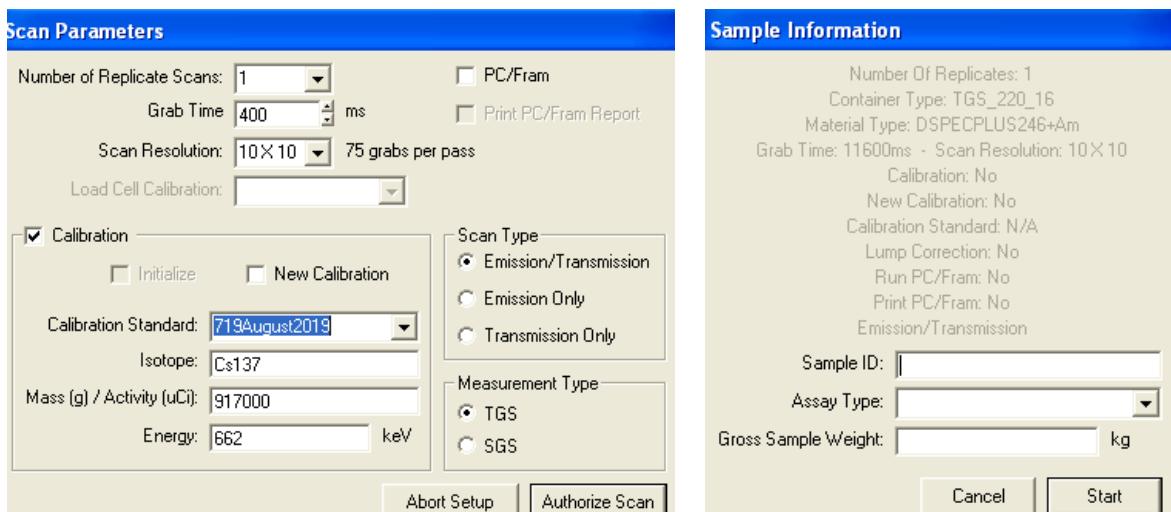


Figure 12 - Scan parameters setting window and Sample Information on *MasterScan* respectively on the left and on the right side (Source: JRC, 2019).

At the end of the process a single calibration measurement for a specific Assay Type and for a specific radionuclide will be produced that will need to go through the analysis process by the *MasterAnalysis* software. In order to have a complete calibration curve for one specific Assay Type this SOP will be repeated for all the radionuclides of interest (e.g. covering all energies range ~ 60 keV to 1.6 MeV).

The analysis of the raw data of the TGS calibration scan will be performed using the GSC and *MasterAnalysis*:

1. From the menu select *File* and *Open* the calibration files (calibration of one file at the time) from the folder: *C:/AntechData/RawData*:
 - a) Click on *ANALYZE*;
 - b) Click *Print Report* to save a PDF report file;
 - c) Click *DONE* to close the procedure and initiate a successive one for the other radionuclides.

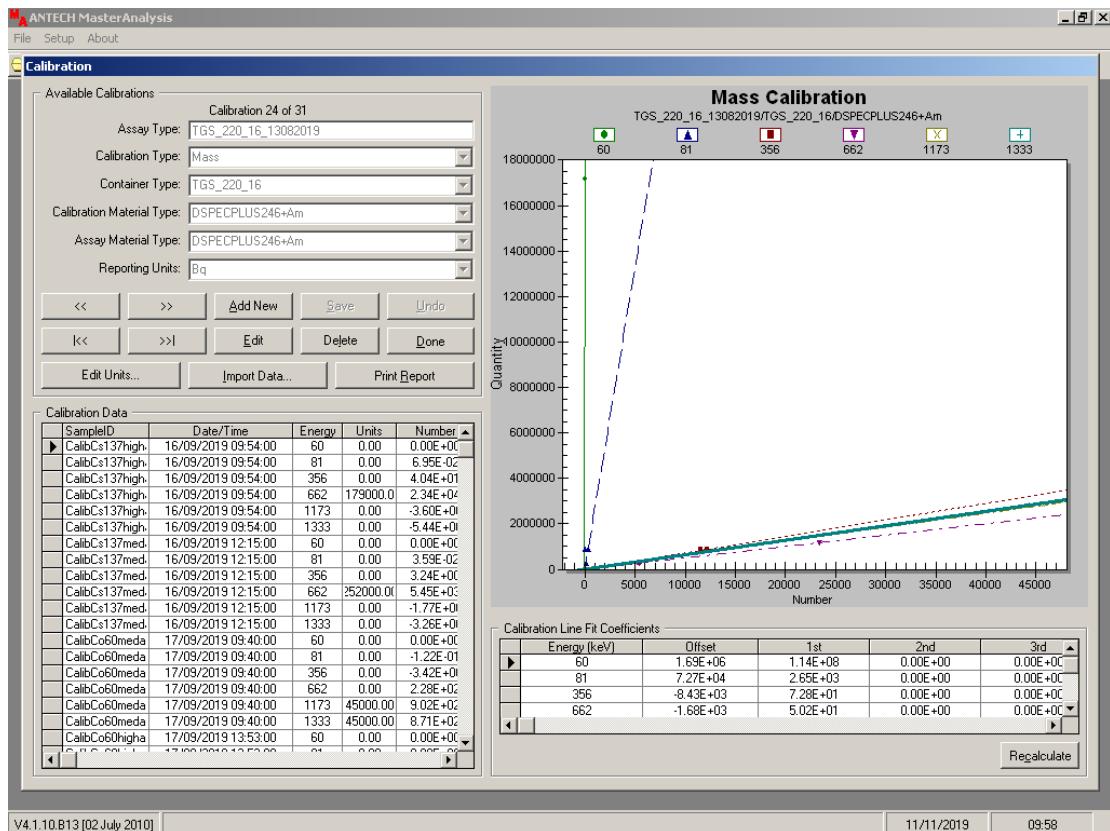


Figure 13 - Mass Calibration for the TGS measurement with four radioactive sources (Source: JRC, 2019).

Configuration

Miscellaneous

| | | |
|--|---|---------|
| 8K TCP/IP Address: | 192.168.13.51 | |
| Machine Type: | DRUM TGS | |
| Machine Name: | WCS-DTGS | |
| Security: | <input type="radio"/> On <input checked="" type="radio"/> Off | |
| Enable Load Cell Measurement: | No | |
| Enable TGS Dead Time Source Measurement: | Yes | |
| Password: | [REDACTED] | |
| Retype Password: | [REDACTED] | |
| Measurement Control Repetitions: | 5 | |
| Length of Straight-through: | 120 | seconds |
| % of Radius to Scan: | 91.2 | |
| Units: | cm, g, kg | |
| Enable Scan Resolution? | Yes | |
| Enable Emission/Transmission Only? | Yes | |
| Display parameters on Sample Information Window? | Yes | |
| Enable User-Defined Text Field on Sample Information Window? | No | |
| Enable User-Defined Option Field on Sample Information Window? | No | |
| Enable Comment Field on Sample Information Window? | No | |
| Enable Tare Weight on Sample Information Window? | No | |
| Append Date/Time to SampleID? | Yes | |
| Enable Daily Straight-Through? | No | |
| Enable Helical SGS Scan? | No | |
| Enable 2-Pass SGS Statistical Test? | No | |

6K Outputs

| | |
|-----------|-------------|
| Output 1: | Amber Light |
| Output 2: | Brake |
| Output 3: | Green Light |
| Output 4: | Shutter |

View Change Log Edit Save Undo Done

Figure 14 - Settings (i.e. Miscellaneous) on the MasterScan program used as REF values for the TSGS SOP (Source: JRC, 2019).

When all the calibration files of interest are analysed, it is necessary to click *Recalculate* on calibration line fit coefficients to let the program to calculate the TGS numbers for all energies (Figure 13).

The reference settings (referred in the next section of the document as REF) of the *MasterScan* program are indicated in the following picture. These are the settings that will be maintained fixed during all validation tests and kept for the SOP.

The following Figure 18 represents the TGS number vs. the Activity for the ^{137}Cs , ^{60}Co , and ^{133}Ba gamma energy lines as evaluated by the *MasterAnalysis* in an empty 220L standard drum at a distance $d = 485\text{ cm}$ from the HPGe detector to the central rotation axis of the drum. Unless specified otherwise, the calibration file (#TGS220_220_16_13082019) was used for all the analysis in this report, in order to derive the activity of unknown radionuclides in a waste drum.

This calibration file (#TGS220_220_16_13082019) is used for all the future analysis in this report, unless it is clearly specified otherwise, in order to derive the activity of unknown radionuclides in a waste drum.

In the calibration the data of ^{241}Am and ^{133}Ba (81 keV gamma line) are not presented because particular consideration has been taken, details of it are discussed in p.50 [16].

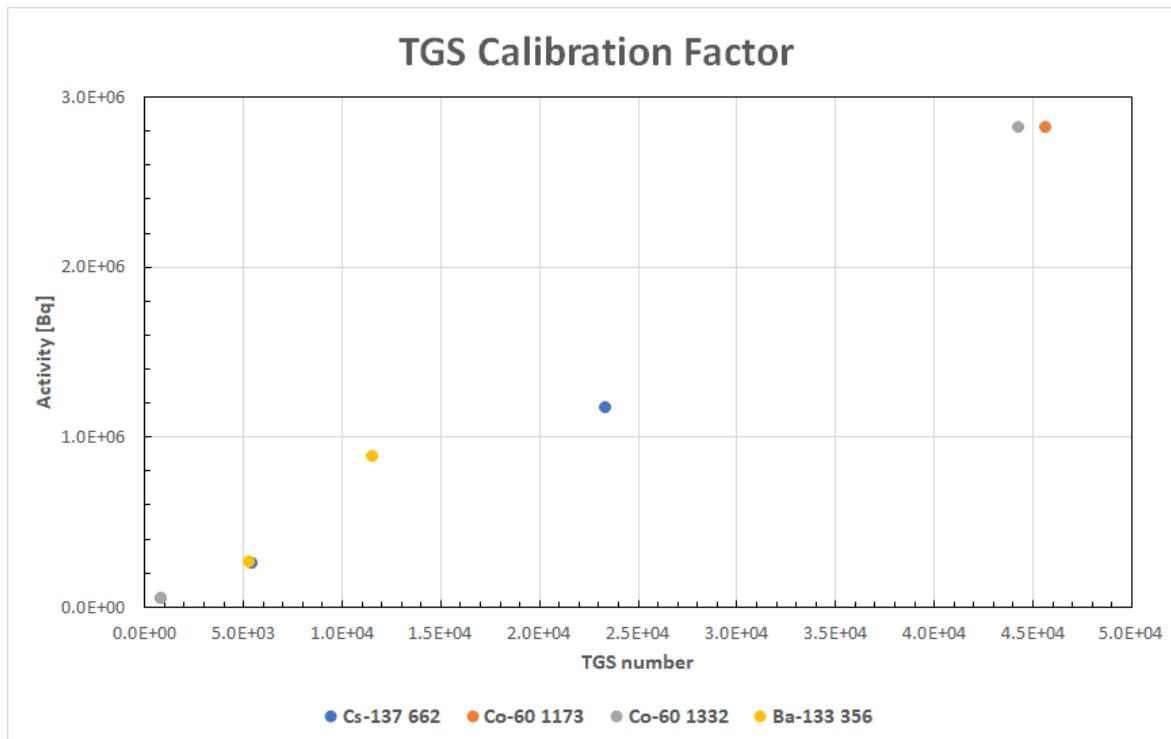


Figure 15 - Calibration curve for ^{133}Ba , ^{137}Cs , and ^{60}Co radioactive sources in an empty standard 220l drum in TGS mode.

9.3.3 Verification of the initial status of the TSGS

Prepare the WCS to perform a TSGS acquisition scan starting from the safety checks, quality control, mass calibration and scan of an unknown sample drum (e.g. 220 L drum).

A PLC panel will present the safety status of the WCS system through gestational software, WCS Process Control (Figure 5) and a LEDs panel in the control room. Following the Standard Operating Procedure (SOP):

Check the FCC, MCC and GSC are switched on and in the same network domain.

Check if the system is in emergency stop status of:

If the indicator *SYSTEM OK* on WCS Process Control is green, the system is not in emergency stop status and the user can perform the measurement.

If the indicator *SYSTEM OK* on WCS Process Control is grey, the system is in emergency stop status and selecting the submenu "WCS Alarms", it is possible to acknowledge the type of alarm/s which produces the emergency stop status.

Solve the emergency stop status by:

- (a) Press the blue button on the Control Instrumentation Panel (LEDs panel);
- (b) Click on **FAULT RESET** button on WCS Process Control and if there are no other faults, the **SYSTEM OK** will confirm the ready status with a green light.

2. The system is ready to perform a TSGS measurement.

9.3.4 Empty matrix drum

The “empty” matrix drum has been used to perform the initial calibration of the TSGS system and for the verification of vertical/channels position independence vs. measured activity. In **Error! Reference source not found.**, we can see a schematic representation of the used vertical position and the 7 channels for the horizontal dependency.

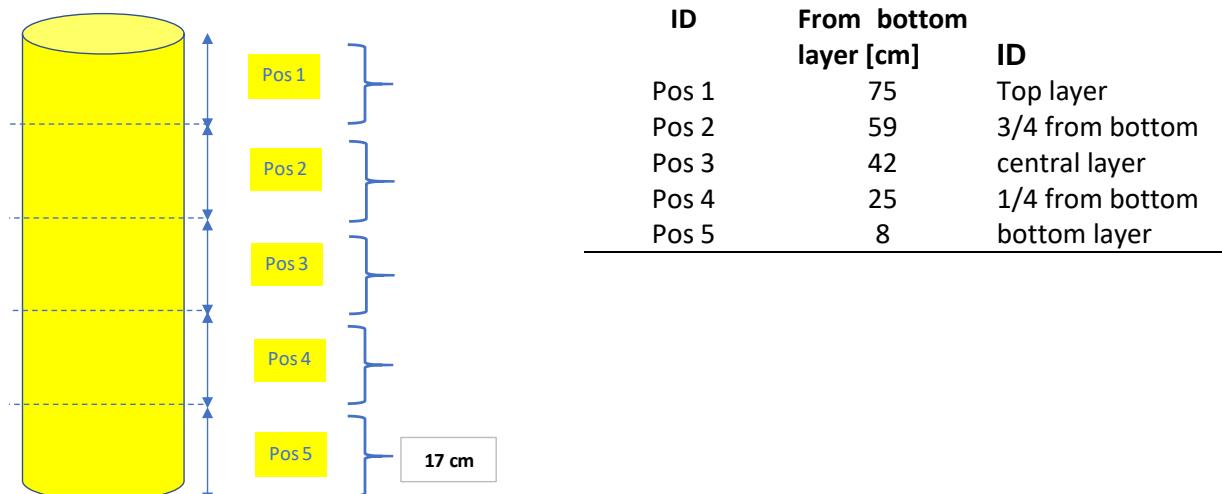


Figure 16 - Schematic representation of the vertical and channel position of a standard radioactive source.

9.3.5 Simulated waste drums SGS

The JRC research team investigated the effect of source position inside a drum performing a series of measurements with ^{137}Cs and ^{60}Co point sources (encapsulated standard source in an Al enclosure) introduced in each of the 5 different matrixes. The standard 220 L drum has been used with radioactive sources introduced in 7 different channels (A to G) and at three different vertical positions (i.e. top, middle and bottom) on the drum (Figure 6) in order to verify the position dependence.

9.3.6 Simulated waste drums TGS

In this section we will go through the SOP for a TGS scan and its relative analysis of a waste drum to determine the activity and hot-spot position of an unknown radionuclide/s. All the steps of this SOP are performed using the GSC. The SOP is the following:

- 1) Put on Gamma station a desired waste drum to be analysed (e.g. a real waste drum or a simulated one with known radioactive content for verification purpose);
- 2) Setup the HPGe detector for bias voltage, gain, shaping time and all related parameters necessary to optimize the electronic detector acquisition chain;
- 3) Open *MasterScan* program and from the menu:
 - a) Select *Setup*;
 - b) Select *Scan Parameters*;
 - c) Define the *grab time* and the scan type, usually is chosen *Emission/Transmission*;
 - d) Click on *AUTHORIZE SCAN* and *RUN*;

- e) Write the name of the measurement in *Sample ID* and the *weight of drum* (adding 22 kg to the gross weight) and choose the *assay type*;
 - f) Click on *START*.
- 4) At the end of measurement open *MasterAnalysis* program and from the menu:
- a) Select File and Open the folder C:/AntechData/RawData;
 - b) Choose the file related to the drum that needs to be analysed;
 - c) Click on *ANALYZE*;
 - d) Move the arrows *Layer* and *Degree* on the right size of the window in order to see the position of the source in the drum [16];
 - e) Write comments if necessary related to the information of the source in the drum on *Post Analysis Comment*;
 - f) Click *Print report* and *DONE* to close. A .pdf report file will be created.

9.3.7 Standard sources position consistency on different matrix drum

Following Table 9 representing the extracted activity data from the Master Analysis program (grab time = 100 s for each layer). In particular, this data represent the activity variation of only a ^{137}Cs source in the 16 layers positioned at the middle height in the Channel A in 220 L drum. The superimposed histogram in Table 9 shows the position consistency activity with the actual setup (higher activity on layer 8).

Table 9 - Measured activity of a ^{137}Cs radioactive source from the MasterAnalysis data extraction in relation to the 16 scanned layers in a TCO 220 L drum (Source: JRC, 2019).

| Somma di Activity | Etichette di colonna | Co60 - 1173 | Cs137 - 662 | Totale complessivo |
|---------------------------|----------------------|------------------|-----------------|--------------------|
| Etichette di riga | | | | |
| Layer 1 | | 69.019 | 78.471 | 402.373 |
| Layer 2 | | 169.67 | 283.575 | 579.772 |
| Layer 3 | | 442.969 | 199.793 | 746.576 |
| Layer 4 | | 563.103 | 591.927 | 1167.671 |
| Layer 5 | | 1825.555 | 2193.296 | 8983.39 |
| Layer 6 | | 5494.099 | 4806.499 | 24856.303 |
| Layer 7 | | 9026.781 | 8106.362 | 42741.876 |
| Layer 8 | | 10048.46 | 9752.414 | 50554.766 |
| Layer 9 | | 8903.538 | 8625.729 | 42716.338 |
| Layer 10 | | 6338.532 | 5477.916 | 27994.931 |
| Layer 11 | | 2477.708 | 2139.641 | 7771.401 |
| Layer 12 | | 659.153 | 813.687 | 1358.021 |
| Layer 13 | | 470.752 | 289.12 | 512.252 |
| Layer 14 | | 199.644 | 213.023 | 313.085 |
| Layer 15 | | 208.793 | 297.323 | 592.351 |
| Layer 16 | | 287.786 | 206.754 | 353.995 |
| Totale complessivo | | 47185.562 | 44075.53 | 211645.101 |
| | | | | 302906.193 |

All the 7 channels have been shown an activity consistency in the vertical position plus a minus one layer (each layer is $\sim 5\text{ cm}$) for a ^{137}Cs and ^{60}Co sources. Extra investigation will be conducted on the extreme positions, top and bottom where the higher vertical differences in comparison to the actual source position have been found for a ^{137}Cs source.

The JRC research team has also investigated the measured activity of a ^{137}Cs source in the three vertical positions (i.e. top, middle and bottom) in different matrix drums. Following is an example of a 220 L “empty” drum where the ^{137}Cs source was positioned respectively in the top, middle and bottom vertical position ($\pm 1\text{ cm}$). It is evident a huge discrepancy between the calculated value of $2.56 + E05\text{ Bq}$ and the measured value in the three positions (i.e. respectively $2.35 + E05$, $2.68 + E05$ and $1.20 + E05\text{ Bq}$) for all channels. The closest value is obtained only in the middle vertical position and in the central “A” channel. In the plot (see Figure 17) are represented with a red, dash blue and dash light blue lines respectively the calculated activity, the negative variation of 10% and the one -20% from the calculated ^{137}Cs activity.

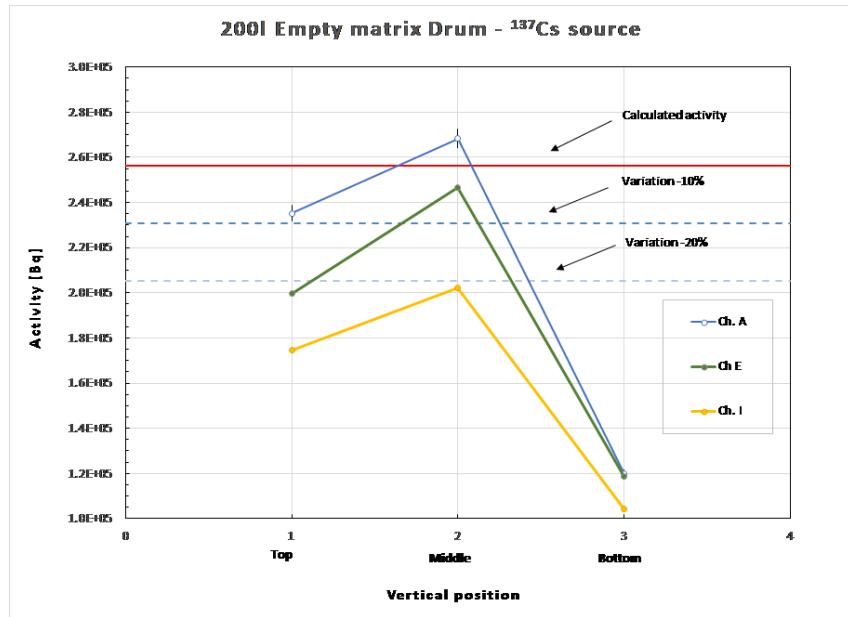


Figure 17 - ^{137}Cs activity variation vs. the vertical position in an empty matrix drum of 220 L (Source: JRC, 2019).

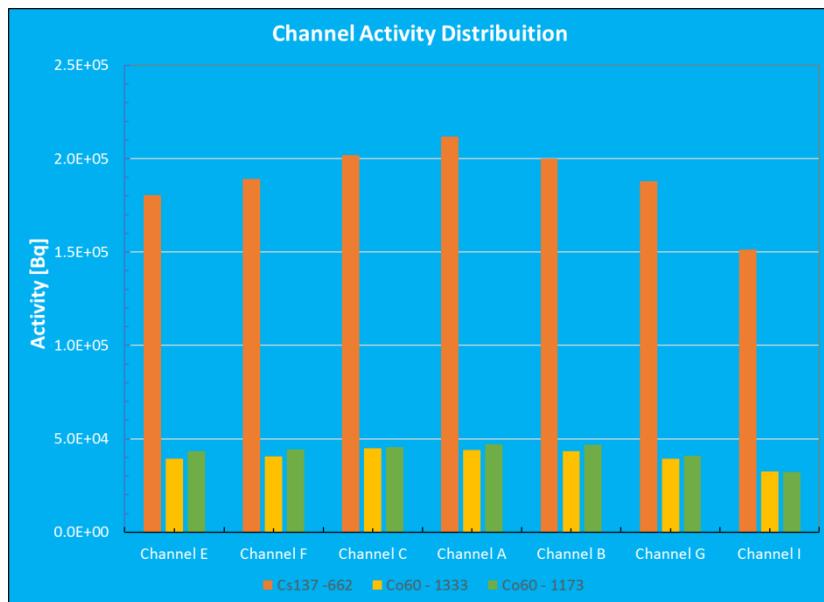


Figure 18 - Activity distribution of ^{137}Cs and ^{60}Co sources in relation to the channel positions (Source: JRC, 2019).

Another set of measurements on simulated waste drums was performed to verify the dependence of all our nuclear vectors (^{133}Ba , ^{137}Cs and ^{60}Co) on the vertical positioning and on the density matrix (TCO, CMT and MTL) respect to the variation of the measured activity vs. the nominal activity.

The summary table shows (Table 10) all the results for ^{133}Ba , ^{137}Cs , and ^{60}Co radioactive sources with names of the files analysed, the activity measured and the difference in percent ($\Delta\%$) between the measured values and the calculated activities at the date of the measurements. Values in Table 10 show a relevant variation on the $\Delta\%$ for all three radionuclides analysed as well for all studied matrices (TCO, CMT and MTL) from a -24% to a -20% for a TCO matrix, from a -80% to a -84% for a CMT matrix and from a -55% to a -71% for a MTL matrix for a standard 1h measurement (aka short measurement time).

Table 10 - Summary of the measurements campaign for the WCS Gamma Station system in TGS mode configuration.

| File ID | Radionuclides | Measured Activity [kBq] | Expected activity [kBq] | Relative deviation [%] |
|--------------------------------------|-------------------|-------------------------|-------------------------|------------------------|
| SHORT MEASUREMENT TIME (~1 h) | | | | |
| TCOoldCs137ChAPo3_16Sep191624 | ¹³⁷ Cs | 1.24E+06 | 1.17E+06 | 5.98 |
| TCOCs137highChAPos1_01Oct190918 | | 8.96E+05 | 1.17E+06 | -23.42 |
| TCOCs137highChAPos4_01Oct191110 | | 8.65E+05 | 1.17E+06 | -26.07 |
| TCOCs137highChIPos4_01Oct191355 | | 9.26E+05 | 1.17E+06 | -20.85 |
| TCOCs137highChIPos3_01Oct191539 | | 9.36E+05 | 1.17E+06 | -20.00 |
| TCOCs137highChGPos3_02Oct190928 | | 9.49E+05 | 1.17E+06 | -18.89 |
| TCOCs137highChAPos5_02Oct191138 | | 9.39E+05 | 1.17E+06 | -19.74 |
| TCOCs137highChGPos2_02Oct191348 | | 9.86E+05 | 1.17E+06 | -15.73 |
| TCOCs137highChCPos3_02Oct191533 | | 9.31E+05 | 1.17E+06 | -20.43 |
| TCOCs137highChAPos2_03Oct190924 | | 9.39E+05 | 1.17E+06 | -19.74 |
| TCOCs137highChCPos2_03Oct191116 | | 9.58E+05 | 1.17E+06 | -18.12 |
| TCOCo60highChAPos2_03Oct191405 | ⁶⁰ Co | 2.13E+06 | 2.75E+06 | -22.55 |
| TCOCo60highChAPos4_03Oct191531 | | 1.90E+06 | 2.75E+06 | -30.91 |
| TCOCo60highChGPos2_04Oct190922 | | 2.32E+06 | 2.75E+06 | -15.64 |
| TCOCo0highChGPos3_04Oct191043 | | 2.20E+06 | 2.75E+06 | -20.00 |
| TCOCo60highChAPos5_04 Oct191205 | | 1.61E+06 | 2.75E+06 | -41.45 |
| TCOCo60highChIPos3_07Oct190915 | | 2.36E+06 | 2.75E+06 | -14.18 |
| TCOCo60highChIPos4_07Oct191037 | | 2.13E+06 | 2.75E+06 | -22.55 |
| TCOCo60highChAPos1_07Oct191201 | | 2.40E+06 | 2.75E+06 | -12.73 |
| TCOoldBa133ChAPo3_18Sep191638 | ¹³³ Ba | 8.57E+05 | 8.85E+05 | -3.16 |
| TCOBa133highChAPos2_07Oct191402 | | 6.57E+05 | 8.82E+05 | -25.49 |
| TCOBa133highChAPos4_07Oct191524 | | 6.39E+05 | 8.82E+05 | -27.57 |
| TCOBa133highChGPos3_08Oct190942 | | 6.69E+05 | 8.82E+05 | -24.12 |
| TCOBa133highChAPos5_08Oct191108 | | 6.98E+05 | 8.82E+05 | -20.82 |
| TCOBa133highChGPos2_08Oct191356 | | 6.58E+05 | 8.82E+05 | -25.43 |
| TCOBa133highChIPos3_08Oct191516 | | 6.30E+05 | 8.82E+05 | -28.59 |
| TCOBa133highChAPos1_10Oct190858 | | 7.83E+05 | 8.82E+05 | -11.18 |
| TCOBa133highChIPos4_09Oct190850 | | 6.25E+05 | 8.82E+05 | -29.14 |
| TCOBa133highChIPos5_09Octo19 | | 6.23E+05 | 8.82E+05 | -29.41 |
| LONG MEASUREMENT TIME (~15 h) | | | | |
| TCOoldCs137ChAPo3_16Sep191624 | ¹³⁷ Cs | 1.24E+06 | 1.17E+06 | 5.98 |
| MTLCs137ChDPos3_26Sep191636 | | 7.83E+05 | 9.14E+05 | -14.33 |
| TCOBa133Cs137Co60_08Oct191649 | | 1.31E+06 | 1.17E+06 | 12.05 |
| TCOCs17Co60Ba133_09Oct191701 | | 1.36E+06 | 1.17E+06 | 16.15 |
| TCOCo60Cs137Ba133_10Oct191658 | | 9.41E+05 | 1.17E+06 | -19.6 |
| MTLCs137Co60Am241Ba1_14Oct191650 | | 4.86E+05 | 9.14E+05 | -46.78 |
| MTLCo60Cs137Am241Ba1_15Oct191653 | | 5.15E+05 | 9.14E+05 | -43.69 |
| CMTCs137Co60Am241Ba1_16Oct191649 | | 6.38E+04 | 1.17E+06 | -94.55 |
| CMTCs137Co60Am241Ba1_16Oct191649 | | 7.68E+05 | 1.17E+06 | -34.40 |
| TCOAm241I2Cs137E4_23Oct191644 | | 1.14E+06 | 1.17E+06 | -2.56 |
| TCOoldCs137ChAPo3_16Sep191624 | | 1.24E+06 | 1.17E+06 | 5.98 |
| MTLCs137ChDPos3_26Sep191636 | | 7.83E+05 | 9.14E+05 | -14.33 |
| TCOBa133Cs137Co60_08Oct191649 | | 1.31E+06 | 1.17E+06 | 12.05 |
| TCOCs17Co60Ba133_09Oct191701 | | 1.36E+06 | 1.17E+06 | 16.15 |
| TCOoldCo60ChAPos3_17Sep191641 | ⁶⁰ Co | 2.84E+06 | 2.78E+06 | 2.16 |

| | | | | |
|----------------------------------|-------------------|----------|----------|--------|
| Co0212metalCanD_25Sep19 | | 1.80E+06 | 2.78E+06 | -35.25 |
| TCOBa133Cs137Co60_08Oct191649 | | 2.78E+06 | 2.75E+06 | 1.09 |
| TCOCs17Co60Ba133_09Oct191701 | | 2.72E+06 | 2.75E+06 | -0.98 |
| TCOCo60Cs137Ba133_10Oct191658 | | 2.77E+06 | 2.75E+06 | 0.73 |
| MTLCs137Co60Am241Ba1_14Oct191650 | | 1.40E+06 | 2.75E+06 | -49.09 |
| MTLCo60Cs137Am241Ba1_15Oct191653 | | 1.82E+06 | 2.75E+06 | -33.82 |
| CMTCs137Co60Am241Ba1_16Oct191649 | | 2.75E+05 | 2.75E+06 | -90.02 |
| CMTCo60Am241Ba133_21Oct191644 | | 1.55E+06 | 2.75E+06 | -43.64 |
| TCOoldCo60ChAPos3_17Sep191641 | | 2.84E+06 | 2.78E+06 | 2.16 |
| Co0212metalCanD_25Sep19 | | 1.80E+06 | 2.78E+06 | -35.25 |
| TCOBa133Cs137Co60_08Oct191649 | | 2.78E+06 | 2.75E+06 | 1.09 |
| TCOCs17Co60Ba133_09Oct191701 | | 2.72E+06 | 2.75E+06 | -0.98 |
| TCOCo60Cs137Ba133_10Oct191658 | | 2.77E+06 | 2.75E+06 | 0.73 |
| TCOoldBa133ChAPo3_18Sep191638 | | 8.57E+05 | 8.85E+05 | -3.16 |
| TCOBa133Cs137Co60_08Oct191649 | | 7.06E+05 | 8.82E+05 | -19.91 |
| TCOCs17Co60Ba133_09Oct191701 | | 8.00E+05 | 8.82E+05 | -9.35 |
| TCOCo60Cs137Ba133_10Oct191658 | | 8.36E+05 | 8.82E+05 | -5.18 |
| MTLCs137Co60Am241Ba1_14Oct191650 | | 5.30E+05 | 8.82E+05 | -39.93 |
| MTLCo60Cs137Am241Ba1_15Oct191653 | | 3.78E+05 | 8.82E+05 | -57.13 |
| CMTCs137Co60Am241Ba1_16Oct191649 | ¹³³ Ba | 5.80E+05 | 8.82E+05 | -34.26 |
| CMTCo60Am241Ba133_21Oct191644 | | 1.42E+04 | 8.82E+05 | -98.39 |
| TCOoldBa133ChAPo3_18Sep191638 | | 8.57E+05 | 8.85E+05 | -3.16 |
| TCOBa133Cs137Co60_08Oct191649 | | 7.06E+05 | 8.82E+05 | -19.91 |
| TCOCs17Co60Ba133_09Oct191701 | | 8.00E+05 | 8.82E+05 | -9.35 |
| TCOCo60Cs137Ba133_10Oct191658 | | 8.36E+05 | 8.82E+05 | -5.18 |
| MTLCs137Co60Am241Ba1_14Oct191650 | | 5.30E+05 | 8.82E+05 | -39.93 |

9.3.8 TGS repeatability

The TSGS allowed us to perform repetitive measurements in order to assess the repeatability of the results. Thus we performed 10 measurements (1400 ms grab time) in a TCO drum with a ^{137}Cs radioactive source ($1.17\text{E}06 \text{ kBq}$) in position A3 (i.e. central channel at a middle height of 220 L drum). In the following plot (Figure 19) our TGS results are showed for the central position in a solid blue line while in red the same results from ANTECH TGS Model 3610 (similar setup conditions with a ^{137}Cs source in the middle centre position [17]), and a dash red line represent the average activity of the 10 measurements in WCS-Ispra.

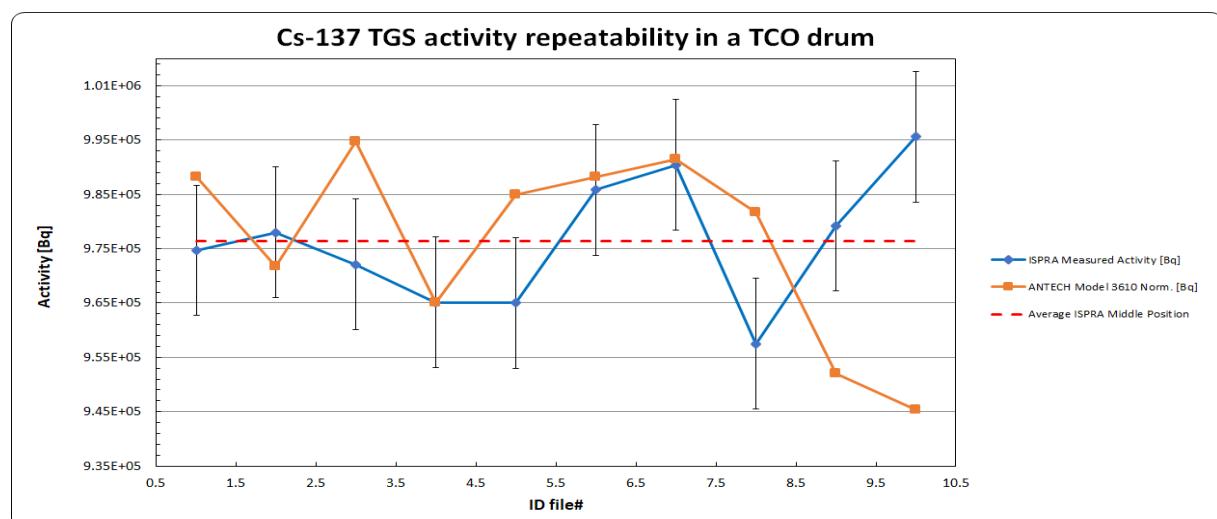


Figure 19 - Ten repetitive measurements in TGS mode with a ^{137}Cs radioactive source in position A3 in a TCO matrix drum.

The Ispra measurement error bars are 1 standard deviation (1.2% of the average value) of the 10 independent measurements because the *MasterAnalysis* was not yet set up to calculate the error on the TGS activity value.

9.3.9 Measurement time dependence

Further analysis in SGS mode (more details on [16]) have shown a dependency on the measurement times (increasing counts) as visible in the following bar graphs of Figure 20 for example in a MTL matrix drum (i.e. with the highest density matrix).

Infact, in the following bar graphs to observe the difference in percent between the calculated activities at the date of the measurement and the measured one in percent variation. The blue indicates short measurement ($\sim 1\text{ h}$) time while the light orange the long measurements ($\sim 15\text{ h}$) time.

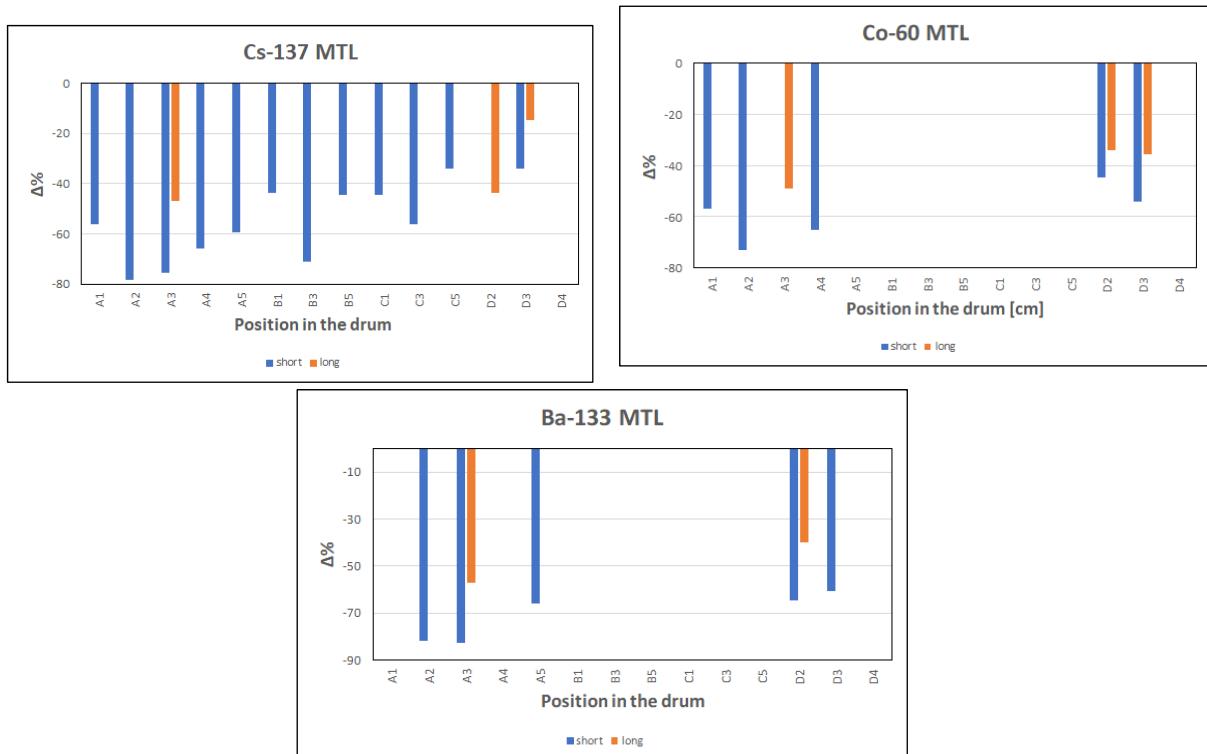


Figure 20 - Activity variation (i.e. ^{137}Cs , ^{60}Co and ^{133}Ba) in percent between short (blue bar) and long (light orange bar) measurement time in a MTL matrix drum.

We have observed (see [16]) that in almost all cases a longer measurement time improves the accuracy of the activities (i.e. lower $\Delta\%$) with different radioactive sources (i.e. ^{137}Cs , ^{60}Co and ^{133}Ba) as well in different matrix drums (TCO, MTL, and CMT), channels and height positions. Those values in Table 10 show variations on the $\Delta\%$ for all three radionuclides analysed as well for all studied matrices (TCO, CMT and MTL) from a 0.8% to a -9% for a TCO matrix, from a -62% to a -66% for a CMT matrix and from a -35% to a -49% for a MTL matrix for an extended 15h measurement (aka long measurement time).

Besides, similar plot reported in [16] revealed that there is a possible dependence on the drum density matrix (TCO<CMT<MTL). It is observable that the activities of the three radionuclides variate from an average discrepancy (i.e. $\langle \Delta\% \rangle$) of 22% to 2% in a TCO, from an average discrepancy of 82% to 64% in a CMT, and from an average of 62% to 41% in a MTL for short time measurements and for long time measurements respectively.

9.3.10 Presence of fissile material

The SGS has been characterized with a CBNM standard of Pu in the central channel at a middle height and in an empty modified calibration matrix drum (see Figure 6 second row). The drum had an aluminium structure to be able to insert the CBNM type samples available at the JRC-Ispra. The holder is constituted by 4 channels distribute as in the picture and by a ‘carrot’, a hollow Al cylinder, that contain the CBNM standard at three possible different vertical positions. From the gamma spectra, it was possible to identify the ^{241}Pu radioisotopes (208.0 keV) and the ^{241}Am gamma line (59.5 keV). In this, way the presence of Pu will be possible be identified also from the Gamma Station and then verified its content with the Neutron Station.

9.3.11 Wastes containing fissile radionuclides

Some analysed TCO drums contain fissile radionuclides: ^{235}U and ^{239}Pu . Their presence has been given by ISOCS which detects ^{238}U and ^{239}Pu from $^{234\text{m}}\text{Pa}$. Since the SGS and TGS calibrations were performed using ^{241}Am , ^{137}Cs , ^{60}Co and ^{133}Ba , the activity of fissile radionuclides is not reported on *MasterAnalysis* but they are detected on the gamma spectrum at the end of each measurements (total emission spectra). The analysed drums containing fissile materials are five (indicated in Figure 6).

At the end of SGS measurement, the acquired spectrum for each drum is reported and the ^{235}U and $^{234\text{m}}\text{Pa}$ - gamma lines are visible at 185.72 keV at 1001 keV. As example, in the following gamma spectra (Figure 21) of the #13456 drum we can observe that they are also visible many of the gamma lines coming from the ^{152}Eu transmission source. Also they are detected many “background” gamma lines (662.7 keV – Cs – 137, 1461 keV – K – 40, 510.8 keV – Th – 232 and many more see Annex II in [16]) as well as far right the artificial digital pulser line at 1975 keV.

For the two gamma lines of interest (^{235}U and $^{234\text{m}}\text{Pa}$) we got 765 ± 59 Net counts and 411 ± 25 Net counts in a TCO matrix drum for Live time = 1531s. If we put the Net counts of those two gamma lines in a table for all the four analyzed drums (see Table 11) we observe that, due to high dead time in the #1510 and #15097 (> 23%), and interfering from other radionuclides in the drums, the $^{234\text{m}}\text{Pa}$ line is not detectable as well as the relative error on Net counts rise on ^{235}U gamma line (> 10%).

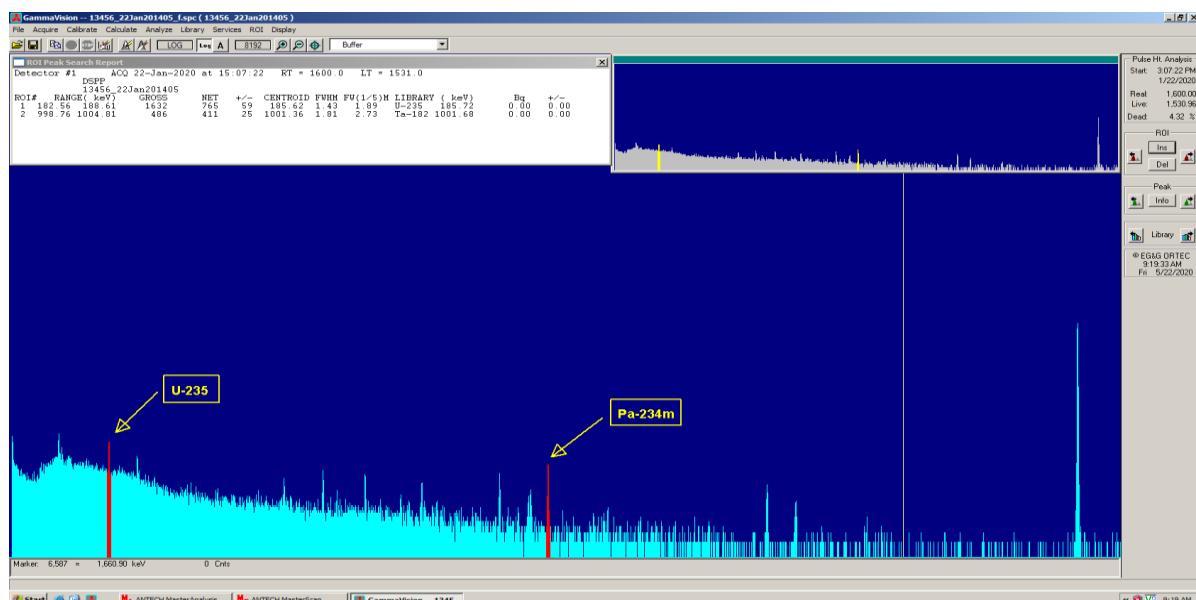


Figure 21 - Gamma spectra with *GammaVision* program showing the U-235 and Pa-234m gamma energy lines for a real waste drum.

Table 11 - Examples from the real waste drums containing fissile materials.

| Ref. number | Radionuclides | Net counts and st. error |
|----------------------|--------------------------------------|--------------------------|
| Real waste drum 1 | ^{235}U (185.7 keV) | 17264 ± 2165 |
| | $^{234\text{m}}\text{Pa}$ (1001 keV) | Not visible |
| Real waste drum 2 | ^{235}U (185.7 keV) | 765 ± 59 |
| | $^{234\text{m}}\text{Pa}$ (1001 keV) | 411 ± 25 |
| Real waste drum 3 | ^{235}U (185.7 keV) | 787 ± 91 |
| | $^{234\text{m}}\text{Pa}$ (1001 keV) | 327 ± 34 |
| Real waste drum 4 | ^{235}U (185.7 keV) | 18325 ± 2149 |
| | $^{234\text{m}}\text{Pa}$ (1001 keV) | 917 ± 68 |

Those considerations on the analysed four spectra for those drums and the fact that we do not have a quantitative evaluation on the activity determined by the *MasterAnalysis* need to be taken as qualitative results (presence or not of fissile materials).

9.3.12 MDA

In this section we will show the calculated MDA values (see eq.(1) [16]) for the three matrices TCO, TNC and RBL) and for the two radionuclides of interest (^{137}Cs and ^{60}Co). As stated in the deliverable A3.1.4 of the MetroDECOM II project: requires an MDA of 0.1Bq/g for ^{137}Cs in a 60kg matrix with a measurement time of 2000s. Particular attention should be put on the different live time and the matrix weight (i.e. 91.4kg for a TCO matrix) of the JRC-Ispra measurements in comparison to the target time A3.1.4. The required uncertainty of 20% cannot be correctly estimated for the JRC measurements where we can only consider a statistical error on the background counts (i.e. 0.5%). In conclusion, the experimental data for ^{137}Cs and ^{60}Co MDA in a TCO, TNC and RBL matrices are showed in the following table (see Table 12). The grey band highlighted the target value of the deliverable A3.1.4 and if we scaled the JRC-Ispra value with the different measurement time we obtain $MDA_{Cs137} = 4\text{Bq} \pm 0.5\%$ while the target value is $MDA_{Cs137} = 6\text{Bq} \pm 20\%$.

Table 12 - MDA calculation for a number of investigated radionuclides for the TSGS system Ispra (highlighted in grey the target MDA for A3.1.4 deliverable closer to JRC measurement conditions).

| Matrix drum | Radioisotope | Energy [keV] | Branching ratio Γ | Abs. Efficiency | Live [s] | time | MDA [Bq/g] |
|----------------|-------------------|--------------|-----------------------------|--------------------|-------------|------|---------------|
| TCO | ^{137}Cs | 662 | 0,85 | 2,04E-04 | 1591 | | 0.038 |
| | ^{60}Co | 1173 | 0,99 | 1,35E-04 | 1569 | | 0.073 |
| TNC | ^{137}Cs | 662 | 0,85 | 2,04E-04 | 4772 | | 0.009 |
| | ^{60}Co | 1173 | 0,99 | 1,35E-04 | 3074 | | 0.003 |
| RBL | ^{137}Cs | 662 | 0,85 | 2,04E-04 | 2305 | | 0.003 |
| | ^{60}Co | 1173 | 0,99 | 1,35E-04 | -- | | NA |

9.3.13 Case of analysis of simulated waste with ^{241}Am content

Particular attention is devoted to study the condition in which a source of ^{241}Am is inside a 220L drum. A set of measurements were performed in different matrix drum (i.e. different density/absorption linear attenuation coefficient) and in different channels for the SGS mode.

Three calibration scans are initialized for the TNC, TCO and RBL matrix drums, and the ^{241}Am source was positioned at different heights and channels. Due to the low activity of the available ^{241}Am point source (e.g. $\sim 17\text{ MBq}$) and the low intensity of the ^{152}Eu transmission source only the TCO drum was analysed [18].

In the Figure 22 the calibration curves for the TCO (that represent the SGS# number vs. the Activity), obtained using ^{241}Am , ^{133}Ba and ^{137}Cs sources, is shown. The two calibrations for ^{241}Am , using two different activities (i.e. $\sim 17\text{ MBq}$ and $\sim 800\text{ kBq}$), are shown in the picture (red circle).

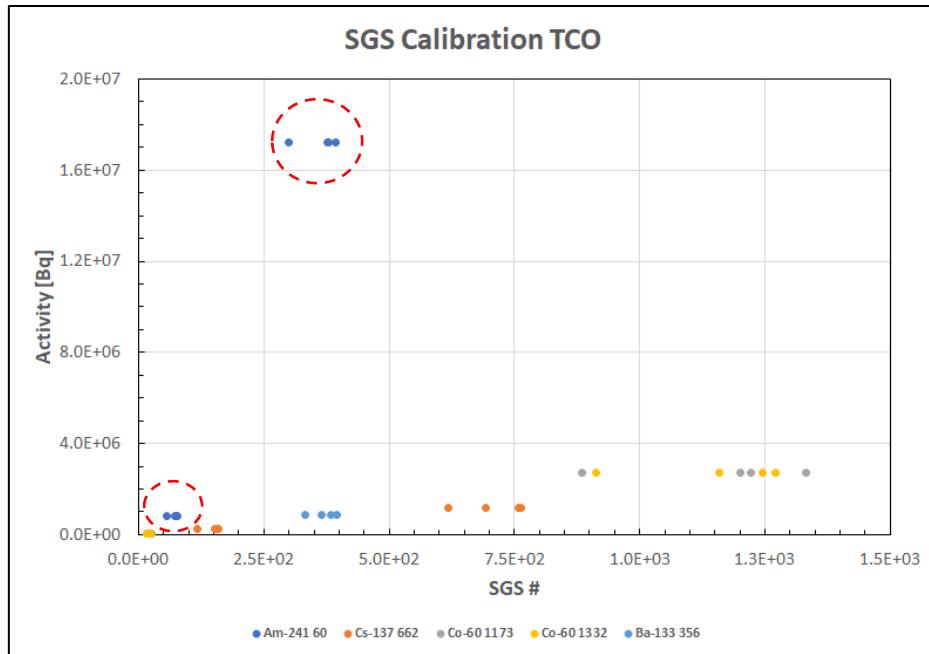


Figure 22 - SGS calibration curves for a TCO matrix drum. Similar for TNC and RBL matrices. Red dashed circles to highlight the two activity used for ^{241}Am .

Besides, the addition of a new calibration point for the ^{241}Am (source of lower activity ^{241}Am of $\sim 800 \text{ kBq}$) has improved the precision of the measured ^{241}Am activity values for all studied matrix (TCO, TNC, RBL) from a 65% to 43% for a TCO to the measured value with a certified method (i.e. In Situ Object Counting System, ISOCS), see [16] and [18].

The SGS was tested using different channel positions (i.e. vertical and horizontal) and grab times (i.e. different time of measurements) to evaluate the statistical accuracy and precision. In Table 13 it is possible to see the different activities calculated by *MasterAnalysis* and the deviation from the actual ^{241}Am activity at the date of the measurement. The first plot (Figure 23) shows the variation of the ^{241}Am activity in relation to the different channel position and with the nominal value (red line) for a TCO drum. The second plot (Figure 24) is the same ^{241}Am source but in relation also to a longer grab time (1600 s) where you have a slightly improvement on the difference between the nominal and the measured activity ($\Delta \sim -4\%$ respect to the $\sim 7\%$ in the shorter measurement) or quite a change for ^{241}Am in TNC drum ($\Delta \sim -14\%$ respect to the $\sim -53\%$ in the shorter measurement). As we observe the different data, we can notice as the measured on external channel positions (like D2 and D4) is 48% and 55% respectively difference from the nominal activity.

Table 13 - Activity measured with 241Am source in a TCO and TNC matrices drum on channel A.

| File ID | Measured Activity [kBq] | Expected activity [kBq] | $\Delta\%$ | Grab time [s] |
|---------|-------------------------|-------------------------|------------|---------------|
|---------|-------------------------|-------------------------|------------|---------------|

| TCO | | | | | |
|----------------------------------|----------|----------|-------|------|--|
| TCOAm241A4Cs137I2_24Oct191642 | 1.64E+07 | 1.72E+07 | -4% | 1600 | |
| TCOAm241C2Cs137G4_25Oct191121 | 1.84E+07 | 1.72E+07 | 7% | 200 | |
| TCOCs137Ba133Co60Am2_29Oct191154 | 1.29E+07 | 1.72E+07 | -25% | 200 | |
| TNC | | | | | |
| TNCAm241ChAPos3_11Nov191106 | 1.71E+07 | 1.72E+07 | -0.5% | 300 | |
| TNCAm241ChCPos4_11Nov191408 | 3.52E+07 | 1.72E+07 | 104% | 225 | |
| TNCAm241ChDPos2_12Nov190958 | 7.61E+07 | 1.72E+07 | 342% | 225 | |
| TNCAm241ChAPos1_12Nov191209 | 1.30E+07 | 1.72E+07 | -24% | 275 | |
| TNCAm241ChCPos2_12Nov191503 | 4.65E+07 | 1.72E+07 | 170% | 175 | |

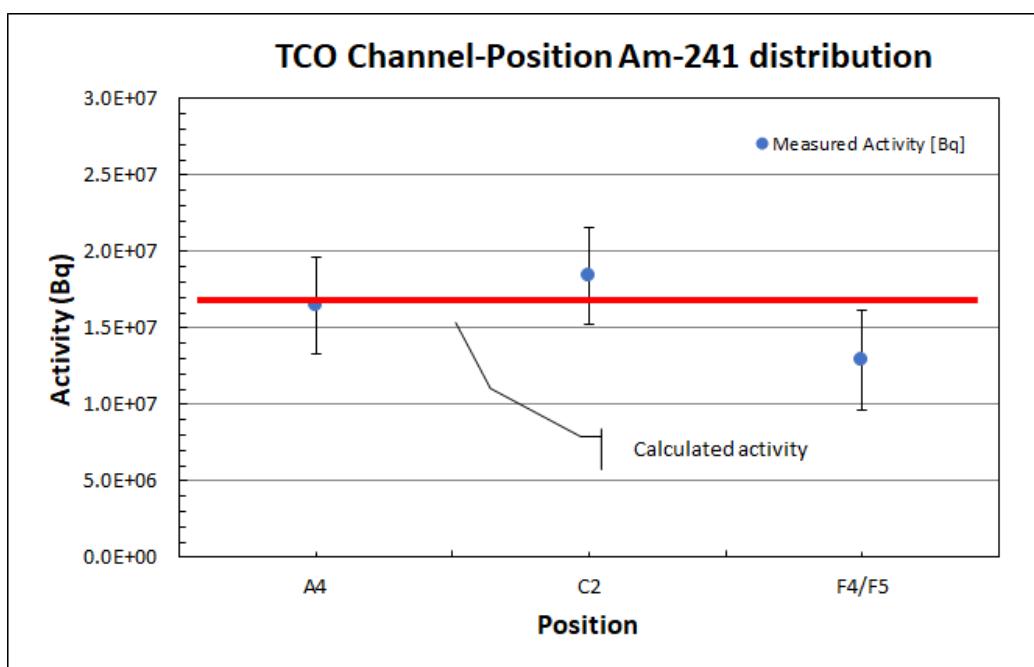
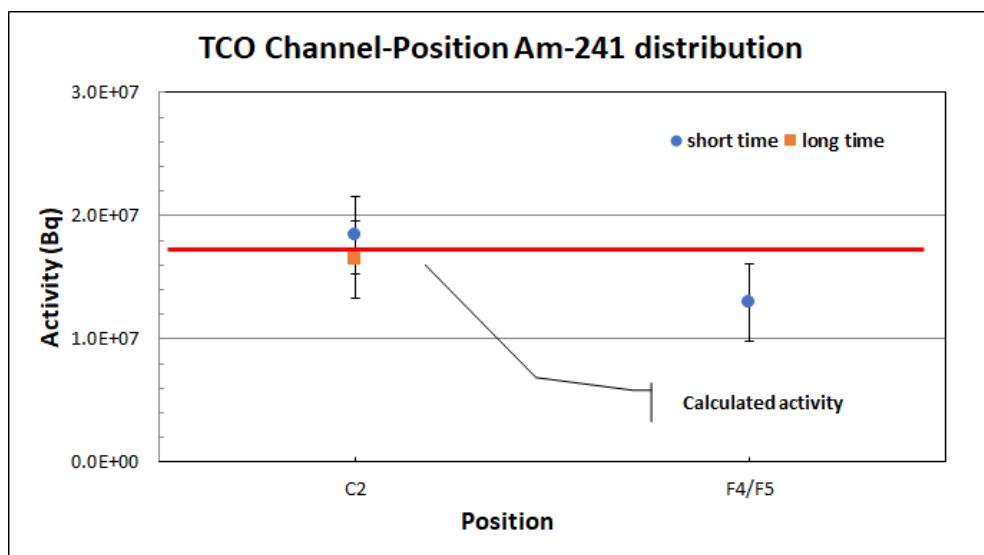
Figure 23 - Distribution of ^{241}Am in different vertical/horizontal position in a TCO drum.

Figure 24 - Distribution of ^{241}Am in different vertical/horizontal position in a TCO drum with different grab time.

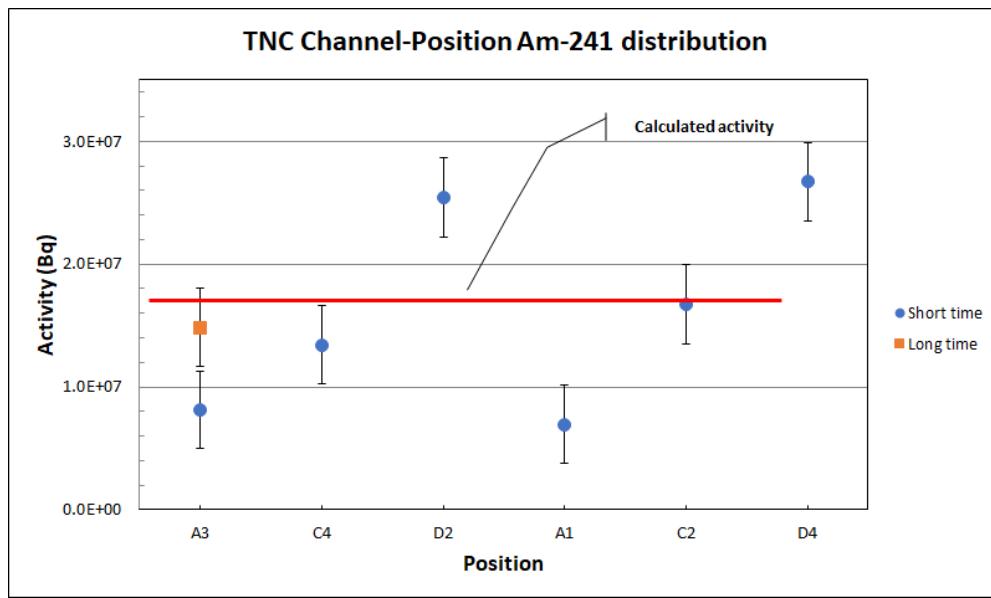


Figure 25 - Distribution of ^{241}Am in different vertical/horizontal position in a TNC drum with different grab time.

9.4 MEASUREMENT CAMPAIGN TSGS

For the measurement campaign, 36 drums have been selected according to the typology of waste matrix, existing ISOCS measurements and radioactive content according to SGS and TGS calibrations.

The selected drums belong to three existing waste matrices: TCO, TNC and RBL. These waste matrices were chosen because at the moment their assay types are well defined and well calibrated for SGS measurements (as reported from [16]). Those three types of matrix delivered by IWMS Consortium represent the wastes stored in the JRC storage building and they are thus produced (IWMS, 2019):

1. Wood, paper and plastic, mixed technological wastes (paper, plastic, chiffonniers or rags, cotton, Tyvek): Technological Waste combustible, **TCO** matrix drum;
2. Glass, fiberglass matrix, glass mixed with paper and plastic: Technological Waste Non-Combustible, **TNC** matrix drum;
3. Demolition wastes, rubbles, stones: Rubbles waste, **RBL** matrix drum.

All the selected drums were produced following to decommissioning and dismantling activities of different buildings at JRC-Ispra site (for more details refer to Table 2 on [16]).

The waste drums have been selected based on their radioactive content measured by ISOCS and reported in WITS (Waste Information Tracking System). The selected drums should contain ^{241}Am , ^{60}Co and ^{137}Cs because only for these radionuclides the calibration measurements have been carried out for SGS and TGS. Besides the choose of them has been based on the target value of MDA (minimum detectable activity) evaluated during the characterisation of the WCS performance in the measurement campaign of 2019 [16].

The selected drums belong to waste categories existing in Area 40 of JRC-Ispra site: VLLW, LLW and ILW. Only two ILW drums were measured and they belong to TCO waste matrix. No ILW drums currently exist at JRC-Ispra site for the TNC and RBL matrix drums.

The SGS measurements were performed according the ISOCS measurement time, usually this value is 1 h but for very old drums the ISOCS measurements was 3 h. Each TGS measurement was performed for 2 h, for more details see [18].

9.5 Results of measurement campaign

The results of 28 representative waste drums are chosen and reported below: 11 for **TCO**, 9 for **TNC** and 8 for **RBL** matrix (Table 14, Table 15, and Table 16, respectively) of which 18 drums were measured by SGS, 12 drums were measured by TGS. These results show the comparison between SGS/TGS and ISOCS measurements for the three radionuclides of interest. In particular the last column of Table 14, Table 15, and Table 16 refers to $\Delta\% = (ISOCS - SGS)/SGS$, in other words the variation of activity from the ANTECH system in SGS mode and the calculated activity from ISOCS system.

Table 14 - Summary of the measurements campaign for the WCS Gamma Station system in SGS and TGS mode configuration for the TCO matrix drum. The green band is for the waste drums containing fissile materials.

| File ID | Radionuclide | ANTECH system Activity [Bq/g] | ISOCS system activity [Bq/g] | $\Delta\%$ |
|-----------------------------|-------------------|---------------------------------------|---------------------------------------|------------|
| 10450TCO_11Dec1912 10 | ^{137}Cs | $3,82\text{E}+00 \pm 5,01\text{E}-02$ | $3,78\text{E}+00 \pm 1,54\text{E}+00$ | 1% |
| | ^{60}Co | $8,95\text{E}-01 \pm 1,67\text{E}-01$ | $8,98\text{E}-01 \pm 3,60\text{E}-01$ | 0% |
| | ^{241}Am | $1,08\text{E}+00 \pm 5,24\text{E}-02$ | $4,16\text{E}-01 \pm 2,78\text{E}-01$ | -61% |
| 14840TCO_11Dec1910 08 | ^{137}Cs | $4,12\text{E}+01 \pm 1,82\text{E}-01$ | $4,33\text{E}+01 \pm 2,03\text{E}+01$ | 5% |
| | ^{60}Co | $6,74\text{E}-01 \pm 1,49\text{E}-01$ | $7,22\text{E}-01 \pm 3,33\text{E}-01$ | 7% |
| | ^{241}Am | $7,62\text{E}+00 \pm 2,04\text{E}-01$ | $1,59\text{E}+00 \pm 1,57\text{E}+00$ | -79% |
| 5194TCO_13Dec19143 5 | ^{137}Cs | $1,63\text{E}+01 \pm 1,99\text{E}-01$ | $1,97\text{E}+01 \pm 1,02\text{E}+01$ | 17% |
| | ^{241}Am | $7,13\text{E}+00 \pm 6,69\text{E}-01$ | $3,90\text{E}+00 \pm 3,63\text{E}+00$ | 83% |
| 5194TCOTGS_28Jan20 0920 | ^{137}Cs | $1,02\text{E}+01 \pm 8,04\text{E}-02$ | $1,97\text{E}+01 \pm 1,02\text{E}+01$ | 93% |
| 15068TCO_13Dec1910 57 | ^{137}Cs | $3,80\text{E}+01 \pm 4,02\text{E}-01$ | $3,49\text{E}+01 \pm 1,69\text{E}+01$ | -8% |
| | ^{60}Co | $3,22\text{E}-01 \pm 1,89\text{E}-01$ | $2,04\text{E}-01 \pm 6,87\text{E}-02$ | -37% |
| | ^{241}Am | $7,00\text{E}+00 \pm 1,63\text{E}-01$ | $2,65\text{E}+00 \pm 2,53\text{E}+00$ | -62% |
| 5187TCO_17Dec19143 0 | ^{137}Cs | $3,74\text{E}+00 \pm 9,26\text{E}-02$ | $3,23\text{E}+00 \pm 1,68\text{E}+00$ | -14% |
| | ^{60}Co | $3,33\text{E}-01 \pm 4,70\text{E}-01$ | $9,03\text{E}-02 \pm 4,41\text{E}-02$ | -73% |
| | ^{241}Am | $7,19\text{E}+00 \pm 1,65\text{E}-01$ | $1,25\text{E}+01 \pm 7,60\text{E}+00$ | 74% |
| 5187TCOTGS_27Jan20 1438 | ^{137}Cs | $3,52\text{E}+00 \pm 8,97\text{E}-02$ | $3,23\text{E}+00 \pm 1,68\text{E}+00$ | -8% |
| 15124TCO_13Dec1909 28 | ^{137}Cs | $1,37\text{E}+02 \pm 1,12\text{E}+01$ | $2,02\text{E}+02 \pm 9,67\text{E}+01$ | 47% |
| | ^{60}Co | $1,23\text{E}-01 \pm 1,80\text{E}-01$ | $1,78\text{E}-02 \pm 1,22\text{E}-02$ | -86% |
| | ^{241}Am | $1,14\text{E}+02 \pm 1,05\text{E}+00$ | $1,89\text{E}+02 \pm 1,78\text{E}+02$ | 66% |
| 15124TCOTGS_28Jan2 01438 | ^{137}Cs | $8,86\text{E}+01 \pm 5,03\text{E}-01$ | $2,02\text{E}+02 \pm 9,67\text{E}+01$ | 128% |
| 15097TCO_22Jan20103 3 | ^{137}Cs | $7,88\text{E}+02 \pm 1,19\text{E}+01$ | $9,39\text{E}+02 \pm 4,41\text{E}+02$ | 19% |
| | ^{60}Co | $2,26\text{E}-01 \pm 1,22\text{E}-01$ | $7,32\text{E}-02 \pm 3,55\text{E}-02$ | -68% |
| | ^{241}Am | $6,90\text{E}+01 \pm 1,41\text{E}+01$ | $6,70\text{E}+01 \pm 6,25\text{E}+01$ | -3% |
| 15101TCO_22Jan20151 0 | ^{137}Cs | $9,21\text{E}+00 \pm 1,07\text{E}-01$ | $9,16\text{E}+00 \pm 4,36\text{E}+00$ | -1% |
| | ^{60}Co | $1,18\text{E}-01 \pm 1,31\text{E}-01$ | $2,43\text{E}-02 \pm 1,33\text{E}-02$ | -79% |
| | ^{241}Am | $7,13\text{E}+00 \pm 6,69\text{E}-01$ | $3,90\text{E}+00 \pm 3,63\text{E}+00$ | -45% |
| 15103_21Jan201528 | ^{137}Cs | $1,21\text{E}+03 \pm 2,32\text{E}+01$ | $1,40\text{E}+03 \pm 6,65\text{E}+02$ | -16% |
| | ^{60}Co | $1,25\text{E}+00 \pm 1,53\text{E}-01$ | $1,49\text{E}+00 \pm 6,95\text{E}-01$ | 19% |
| | ^{241}Am | $1,08\text{E}+02 \pm 7,99\text{E}+00$ | $4,65\text{E}+01 \pm 4,31\text{E}+01$ | -57% |

Table 15 - Summary of the measurements campaign for the WCS Gamma Station system in SGS and TGS mode configuration for the TNC matrix drum.

| File ID | Radionuclide | ANTECH system Activity [Bq/g] | ISOCS system activity [Bq/g] | $\Delta\%$ |
|---------|--------------|-------------------------------|------------------------------|------------|
|---------|--------------|-------------------------------|------------------------------|------------|

| File ID | Radionuclide | ANTECH system Activity [Bq/g] | ISOCS system activity [Bq/g] | $\Delta\%$ |
|--------------------------|-------------------|---------------------------------------|---------------------------------------|------------|
| 3383TNC_22Jan201150 | ^{137}Cs | $4,15\text{E}+00 \pm 3,62\text{E}-02$ | $5,35\text{E}+00 \pm 2,78\text{E}+00$ | 29% |
| | ^{241}Am | $3,50\text{E}+00 \pm 2,14\text{E}-01$ | $2,70\text{E}+00 \pm 1,75\text{E}+00$ | -23% |
| 3383TNCTGS_30Jan201151 | ^{137}Cs | $2,68\text{E}+00 \pm 6,74\text{E}-02$ | $5,35\text{E}+00 \pm 2,78\text{E}+00$ | 50% |
| 12370TNC_23Jan200910 | ^{137}Cs | $3,28\text{E}+01 \pm 3,92\text{E}-01$ | $4,60\text{E}+01 \pm 2,19\text{E}+01$ | 40% |
| | ^{60}Co | $6,70\text{E}-02 \pm 9,18\text{E}-03$ | $1,37\text{E}-01 \pm 4,69\text{E}-02$ | 104 % |
| | ^{241}Am | $4,89\text{E}+00 \pm 1,30\text{E}-01$ | $5,85\text{E}+00 \pm 5,43\text{E}+00$ | 20% |
| 12370TNCTGS_29Jan201206 | ^{137}Cs | $1,99\text{E}+01 \pm 5,57\text{E}-01$ | $4,60\text{E}+01 \pm 2,19\text{E}+01$ | 131 % |
| | ^{60}Co | $5,06\text{E}-02 \pm 3,27\text{E}-03$ | $1,37\text{E}-01 \pm 4,69\text{E}-02$ | 171 % |
| 13516TNC_23Jan201020 | ^{137}Cs | $5,01\text{E}-01 \pm 1,98\text{E}-02$ | $4,06\text{E}-01 \pm 1,61\text{E}-01$ | -19% |
| | ^{241}Am | $3,97\text{E}+00 \pm 1,45\text{E}-01$ | $3,96\text{E}+00 \pm 1,61\text{E}+00$ | - 0,3% |
| 13516TNCTGS_29Jan201418 | ^{137}Cs | $3,68\text{E}-01 \pm 1,85\text{E}-03$ | $4,06\text{E}-01 \pm 1,61\text{E}-01$ | 9% |
| 13513TNC_24Jan200917 | ^{137}Cs | $1,03\text{E}+00 \pm 2,29\text{E}-02$ | $8,56\text{E}-01 \pm 3,42\text{E}-01$ | -17% |
| | ^{241}Am | $2,55\text{E}+00 \pm 1,27\text{E}-01$ | $3,89\text{E}+00 \pm 1,61\text{E}+00$ | 49% |
| 13513TNCTGS_30Jan2010938 | ^{137}Cs | $7,73\text{E}-01 \pm 9,11\text{E}-03$ | $8,56\text{E}-01 \pm 3,42\text{E}-01$ | 10% |
| 13585TNC_23Jan201417 | ^{137}Cs | $5,80\text{E}+00 \pm 7,52\text{E}-03$ | $4,10\text{E}+00 \pm 1,68\text{E}+00$ | -29% |
| | ^{241}Am | $1,30\text{E}+00 \pm 1,78\text{E}-01$ | $2,47\text{E}-01 \pm 0,00\text{E}+00$ | -81% |
| 13585TNCTGS_30Jan201412 | ^{137}Cs | $3,63\text{E}+00 \pm 6,89\text{E}-02$ | $4,10\text{E}+00 \pm 1,68\text{E}+00$ | 13% |

Table 16 - Summary of the measurements campaign for the WCS Gamma Station system in SGS and TGS mode configuration for the RBL matrix drum.

| File ID | Radionuclide | ANTECH system Activity [Bq/g] | ISOCS system activity [Bq/g] | $\Delta\%$ |
|-------------------------|-------------------|---------------------------------------|---------------------------------------|------------|
| 1785_24Feb201516 | ^{137}Cs | $4,51\text{E}+02 \pm 6,97\text{E}-01$ | $1,82\text{E}+02 \pm 7,20\text{E}+01$ | -60% |
| | ^{60}Co | $2,72\text{E}+00 \pm 1,67\text{E}-02$ | $1,44\text{E}+00 \pm 5,92\text{E}-01$ | -47% |
| | ^{241}Am | $2,98\text{E}+01 \pm 2,44\text{E}-01$ | $2,28\text{E}+00 \pm 0,00\text{E}+00$ | -92% |
| 1785TGSRBL_28Feb201359 | ^{137}Cs | $3,00\text{E}+02 \pm 1,22\text{E}+01$ | $1,82\text{E}+02 \pm 7,20\text{E}+01$ | -39% |
| | ^{60}Co | $2,12\text{E}+00 \pm 9,97\text{E}-02$ | $1,44\text{E}+00 \pm 5,92\text{E}-01$ | -32% |
| 3299_24Feb201018 | ^{137}Cs | $1,38\text{E}+02 \pm 5,28\text{E}-01$ | $9,79\text{E}+01 \pm 3,94\text{E}+01$ | -29% |
| | ^{60}Co | $1,12\text{E}-01 \pm 1,03\text{E}-02$ | $8,50\text{E}-02 \pm 3,50\text{E}-02$ | -24% |
| | ^{241}Am | $1,10\text{E}+01 \pm 3,42\text{E}-01$ | $6,73\text{E}-01 \pm 5,70\text{E}-01$ | -94% |
| 14748_24Feb201401 | ^{137}Cs | $2,76\text{E}+02 \pm 5,49\text{E}-01$ | $2,05\text{E}+02 \pm 8,29\text{E}+01$ | -26% |
| | ^{241}Am | $3,30\text{E}+01 \pm 2,82\text{E}-01$ | $7,19\text{E}+00 \pm 3,04\text{E}+00$ | -78% |
| 14748TGSRBL_28Feb201132 | ^{137}Cs | $2,83\text{E}+02 \pm 7,12\text{E}+00$ | $2,05\text{E}+02 \pm 8,29\text{E}+01$ | 38% |
| 14749_26Feb201352 | ^{137}Cs | $4,93\text{E}+01 \pm 1,51\text{E}-01$ | $3,40\text{E}+01 \pm 1,38\text{E}+01$ | -31% |
| | ^{241}Am | $5,09\text{E}+00 \pm 2,64\text{E}-01$ | $2,28\text{E}+00 \pm 1,49\text{E}+00$ | -55% |
| 14749TGSRBL_27Feb200954 | ^{137}Cs | $5,15\text{E}+01 \pm 2,28\text{E}+00$ | $3,40\text{E}+01 \pm 1,38\text{E}+01$ | -34% |
| 14798_26Feb201149 | ^{137}Cs | $1,40\text{E}+00 \pm 4,30\text{E}-02$ | $6,10\text{E}-01 \pm 2,45\text{E}-01$ | -56% |
| | ^{241}Am | $8,51\text{E}-01 \pm 9,62\text{E}-02$ | $1,07\text{E}-01 \pm 0,00\text{E}+00$ | -87% |
| 14798TGSRBL_27Feb201206 | ^{137}Cs | $1,05\text{E}+00 \pm 7,01\text{E}-02$ | $6,10\text{E}-01 \pm 2,45\text{E}-01$ | 42% |

The full table with all 53 analysed drums in SGS and TGS mode is available in [18] for more details as well as a pivot table of the data distribution in relation to the SGS/TGS mode, VLLW, LLW and ILW, and matrix type.

10 VALIDATION OF GAMMA STATION (TGS)

The TGS extends the range of gamma-ray measurement technology, as it is able to determine the radionuclide inventory in heterogeneous matrices. The TGS has the capability to generate images of the distribution of both absorbers and radionuclides within drums.

In TGS mode, the system will perform a rotation and a horizontal movement of the drum with respect to the gamma transmission source axis. It will acquire 150 separate spectra while the system will create a helicoidally drum movements (i.e. translation and rotation). In this tomography process, after a geometric transformation of the different coordinate system, the Master Analysis will compute a distribution of the activity and an attenuation coefficient map in a rectangular grid for each scanned layers. Through the attenuation map, the operator is able to visualize the variation of density in different regions of the drum. The TGS will perform a two-pass scanning to produce emission data and transmission data for attenuation correction, but voxel by voxel and then segment by segment. The TGS extends the capability of the WCS because it is able to correct the emission data with the attenuation coefficient in a heterogeneous matrix.

As in the previous sections (§9.1.1-9.1.2), the TGS system used the same components (hardware and software) of the SGS system. The only difference is the shape of the collimator that it must be the diamond shape.

10.1 MEASUREMENT CAMPAIGN

From those standard drum matrixes the following three are used for our real waste measurements:

1. Technological waste, combustible (TCO)
2. Technological waste, non-combustible (TNC)
3. Rubbles waste (RBL)

Results of the measurement campaign for TGS are reported in the previous §9.5 together with SGS results to address the ISO/IEC 17025:2017 standard (§7.3.2 for b), d), e) and g) and §7.3.3 for a), b), c), d), e), f), h), i)).

11 VALIDATION OF PASSIVE NEUTRON STATION

11.1 SYSTEM DESCRIPTION

The passive/active neutron station is designed and optimised to accommodate standard waste drums up to the size of 220 L. Drum transfers in and out of the central cavity is done automatically by means of a sturdy cantilever mechanism. In Figure 26 the loading beams of the cantilever can be seen located in front of the open door of the neutron station. The motor-operated door of the station constitutes one entire side of the instrument. When fully opened one side of the parallelepiped shaped cavity is completely accessible from that side, see Figure 26 below.



Figure 26 - Mechanical structures of the passive/active neutron station.

The neutron station is constructed using a load-bearing plinth coupled to an upper material support frame. Internal structural members are constructed from aluminium alloy and external members from steel. The structure has a base plinth that permits the whole measurement structure to be jacked and aligned with the loading conveyor (located in front of the door, parallel with the door).

The mechanical structure provides support for:

1. the measurement cavity graphite layer
 2. the measurement cavity polythene layer
 3. a rotating drum support (with incisions for drum transfer by cantilever beams)
 4. support racks for installation and maintenance of ${}^3\text{He}$ detector tubes, head amplifiers and signal cabling loom
 5. in-cavity instrumentation including neutron flux and source monitors.
- b)

During operation the measurement cavity is covered by a large sliding door. The door is moved on floor mounted linear slides by a ball screw actuator. The door incorporates a through beam safety light curtain and bar code reader (to identify the drum as it enters the chamber).

A cross section of the passive/active neutron station based on the original design drawings is shown in Figure 27 [19].

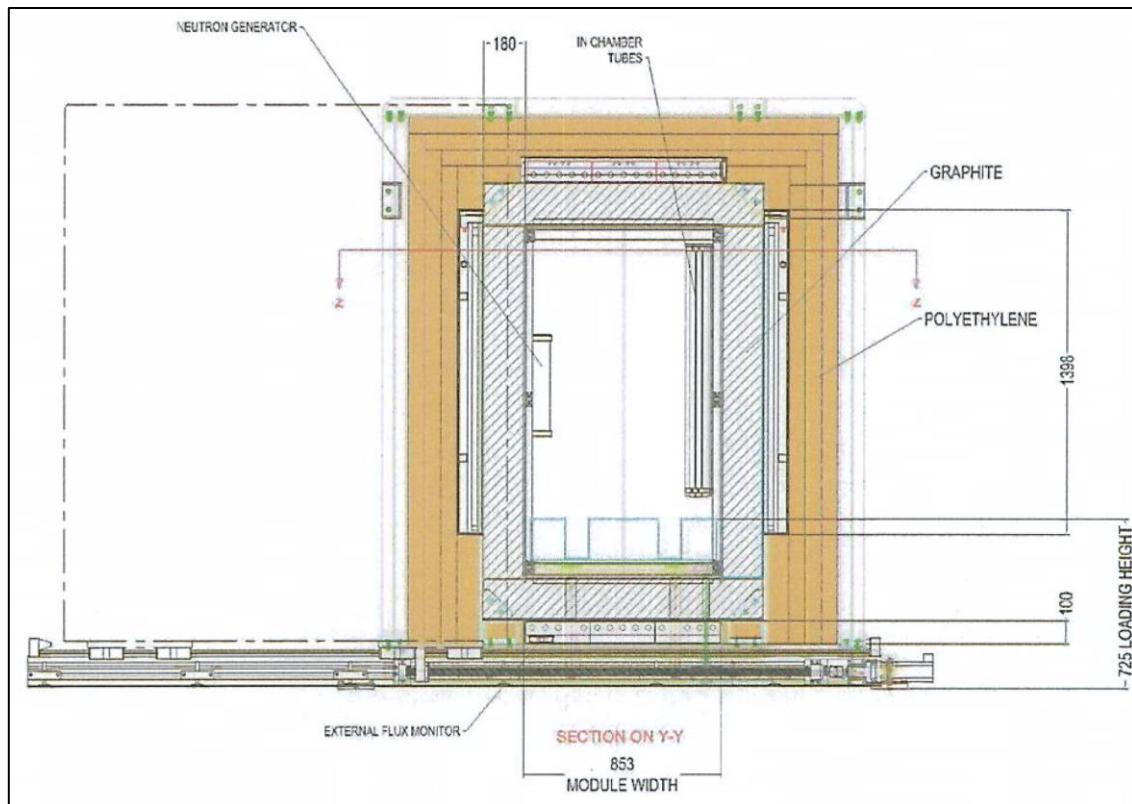


Figure 27 - Sketch of internal components of the passive/active neutron station.

All six sides surrounding the central cavity have the same material composition in the following order (from external to internal) including:

- An external metal skin to reduce the risk of fire and to enable easy decontamination if needed.
- A substantial polyethylene shield (3 x 70 mm thick) to reduce the neutron background detection in the ^3He modules, and to reduce the radiation dose outside the station when the neutron generator is in use.
- Neutron detector modules composed of cadmium lined polyethylene modules with ^3He detector tubes embedded. The width of the modules is 70 mm. The modules and the ^3He tubes cover the full volume of the drum cavity on all sides. The active length of the vertical ^3He tubes is 1300 mm. The total number of ^3He tubes is 88 (15 on each side with 2 tubes removed on bottom to make space for mechanical supports).
- A graphite liner of 180 mm thickness completely covering the drum cavity on all six sides. The graphite is nuclear grade with neutron absorber impurities removed.
- An internal aluminium skin covers the cavity completely for easy decontamination if necessary.

The volume of the central cavity is approximately 850 mm by 850 mm (horizontal cross section) by 1300 mm height (above drum base plinth). This central volume is sufficient to accommodate a standard waste drum up to 400 L, the neutron generator and some flux monitoring neutron detectors. The drum plinth has in addition a turntable mechanism able to rotate the drum during measurement if this is considered advantageous for the data acquisition.

Although the station can accommodate standard drums up to 400 L, the instrument is optimised for measurement of 220 L drums. The waste drums to be measured are limited to maximum 1,500 kg. The maximum expected density is 400 kg/m^3 with a typical range from 300 to $2,500 \text{ kg/m}^3$. The waste matrices may be composed of low-density process waste material, super-compacted waste, bitumen, cement or metal.

The conveyor and the cantilever subsystem are capable of automatically delivering, loading and unloading drums up to 1500 kg.

A neutron generator is used as the external neutron source when the passive/active neutron station is operated in active interrogation mode.

The Passive/active neutron station and all access doors to the measurement hall of the building, are equipped with safety interlocks firstly to avoid that the neutron generator can be switched on before the station is completely closed and the WCS room is completely evacuated, and secondly to reduce the risk of accidents caused by moving parts of the station (door, rotating plinth and cantilever).

11.2 SYSTEM CHARACTERISTICS

The passive neutron counting measurement system of WCS passive/active neutron station has been described in ref [10] and [20]. The principle scheme for the neutron detection and signal acquisition system is also shown in Figure 28. A total of eighteen neutron detection modules surround the central drum cavity. Each module is composed of five ${}^3\text{He}$ gas proportional counters embedded in polyethylene. The front-end electronics of each module has a signal pre-amplifier, amplifier and discriminator circuit (named JAB-01) producing a TTL logic pulse for each neutron detection event.

During the initial testing of the neutron detection system, some imperfections that would affect the measurement results were uncovered. Most importantly the JAB-01 amplifier systems, although very robust and compact, have characteristic (such as very short signal shaping time of less than 200 ns) which make the system prone to occasionally producing false detection events (double events). In waste assay systems, where counting rates are relatively small (in contrast to safeguards systems for bulk materials where counting rates are high), this is a particular problem. In addition, the application of the neutron correlation analysis [21], [22], where signal multiplets are used, the disturbance of double pulses can be devastating. Also other kinds of electronic noise are often seen in large detections systems with many parallel detection chains. The resources available in the present project did not allow for a complete revision of the detection system of the passive/active neutron station. For this reason it was decided to reduce the electronic noise by digital filtering.

The filtering of the digital signal pulse train was conducted in the Time Digitizer for Safeguards (TDS32) unit shown in Figure 28. The TDS32 was re-programmed to not only merge the eighteen signal inputs into one pulse train, but also to disallow any signal occurring less than 1 μs after a given signal. In addition, if the following signal would occur on an input line different from the leading signal, the “dead-time” period would be extended by another period of 1 μs . Clearly this rather crude mechanics for filtering digital noise signals is only acceptable at low counting rates (as in waste assay). The filtering effect of the final pulse train, fed into the Canberra JSR-14 correlation analyser, was however very positive and indeed necessary for achieving acceptable results.

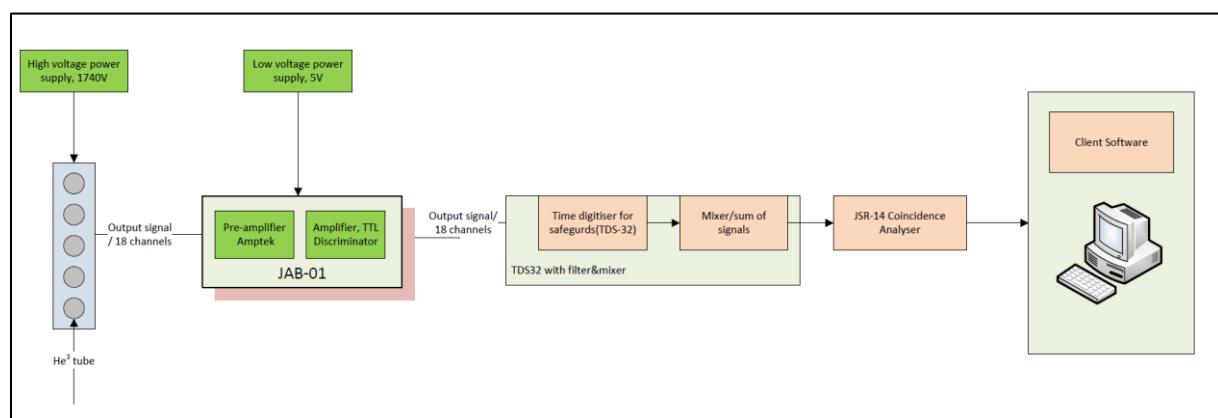


Figure 28 - Neutron counting data acquisition system.

The TDS32 designed and built at JRC has 32 digital input channels. The device is housed in a standard 1U 14-inch rack enclosure. The device can utilize a USB3.0 data link capable of a real data throughput of 300 MB/s of list-mode digital signals. The front panel has a row of Light Emitting Diodes (LED) to let the user observe the presence of input pulses on each line. The device is powered through a 230 V power cord. The TDS32 includes a data acquisition application software programmed in Python, featuring a graphical user-interface (GUI). The device also performs the digital signal mixing mentioned above in FPGA hardware to output the pulse train to the signal analyser. Filtering of the pulse train using the Moving Window Deconvolution (MWD) algorithm, or other digital filtering algorithms, can substitute the classical analogue processing chain even at higher data acquisition rates, including the implementation of the shift-register analysis.

11.2.1 System characteristics

As the passive/active neutron station is intended for both passive and active neutron measurements the design, as described above, is ultimately a compromise to accommodate both methods. In favour of the passive measurements, the instrument has relatively many ^3He detectors embedded in rectangular polyethylene modules. This benefits greatly the detection of so-called coincident neutrons, or correlated neutrons, from spontaneous fission as the key measured quantity in the passive neutron method. All ^3He detector modules are covered in a cadmium liner on all sides thus reducing the neutron flux coupling between the waste matrix and the detector modules. This feature renders the average neutron life-time in the detection modules (relatively) independent of the neutron absorption properties of the waste matrix in a given drum, although at the expense of a reduction in the fission neutron detection efficiency. The average thermal neutron life-time, like the neutron detection efficiency, is an important parameter in the passive neutron coincidence technique.

The size (thickness) of the graphite mantle is a design compromise aiming at achieving both a high interrogating thermal neutron flux in the cavity, and a (sufficiently) good detection efficiency for fission neutrons in the ^3He detector modules located outside the graphite mantle.

The passive neutron measurement is intended to determine the content of spontaneous fissile isotopes such as the even mass number plutonium isotopes (^{238}Pu , ^{240}Pu and ^{242}Pu) by measurement of the neutron emission from the waste package. If the waste contains unprocessed spent fuel, spontaneous fission is typically dominated by the ^{244}Cm which may significantly disturb the passive neutron determination of plutonium. Besides the spontaneous fission neutrons, neutrons may be emitted from the waste from (α, n) reactions if for example PuO_2 or $^{241}\text{AmO}_2$ is present in the waste.

In order to isolate the spontaneous fission neutron detections from (α, n) neutron detections the so-called neutron correlation technique, or neutron coincidence counting, is applied. This is a standard nuclear safeguards technique applied for mass determination of plutonium. The main difference between safeguards measurements of bulk Pu samples and Pu in waste is that for the latter the neutron detection efficiency of the instrument cannot be assumed due to the absorption of neutrons in the waste matrix. For this reason setting up of instrumentation for waste assay by neutron detection, requires careful calibration measurements of standardised artificial waste matrices and known Pu standards.

In the passive neutron correlation technique [23], a so-called Shift Register is applied to record the time sequence of neutron detection events in the primary detection system (the fast neutron detection modules located outside the graphite mantle). By observing pairs of neutrons originating from spontaneous fission events, the spontaneous fission rate in the drum can be determined. This is done to eliminate the contribution of single neutrons (uncorrelated neutrons) produced by (α, n) reactions. Determining the spontaneous fission rate, and by knowledge of the Pu isotopic composition of the waste package (e.g. by gamma spectroscopy), the total Pu mass can be derived.

11.2.2 Neutron detection system

Prior to launching the measurements with neutron sources, significant time was spent on the setup and testing of the neutron detection system.

The 18 fast neutron detector modules of the passive/active neutron station each has a pre-amplifier/amplifier/discriminator circuitry producing a TTL signal for each detection neutron. The analogue electronics is enclosed in a sealed junction box. Each individual electronic chain is connected to five ${}^3\text{He}$ detectors was tested to yield the best signal/noise ratio for a common high voltage setting. The final configuration (amplifier gain, discriminator setting) was the best compromise for all modules when operated with a common HV of 1500 volt applied to all ${}^3\text{He}$ detector tubes.

We measured the die-away time of the passive/active neutron station using a ${}^{252}\text{Cf}$ source placed in the centre of the otherwise empty cavity. Data were acquired with a standard Multi Channel Scalar (MCS), and the measurement points fitted to a single exponential decay. See Figure 29.

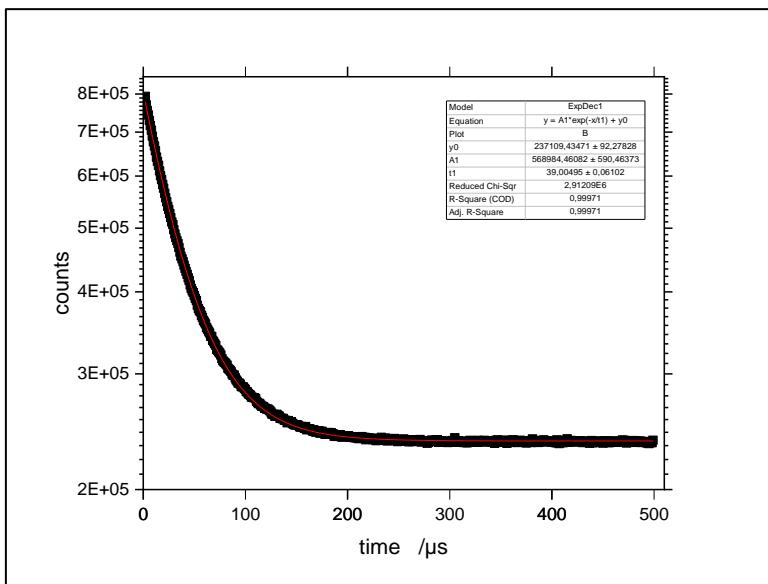


Figure 29 - Number of neutron detections as function of time following a neutron detection event at time zero (MCS histogram), including fitting parameters to a single exponential fit.

The least-squares fitting to a single exponential curve is indeed very good. The fitting parameter, the die-away time, equals to $t_1 = 39.00 \pm 0.06 \mu\text{s}$. The very good fit indicates a good geometrical design of the fast neutron detection system in the passive/active neutron station.

The neutron detection efficiency is a key parameter for a passive neutron detection system. The efficiency of the passive/active neutron station was measured to 12.24 % with a ${}^{252}\text{Cf}$ source centred in an empty drum.

The efficiency, and the efficiency profile (efficiency as function of source location), are important parameters in waste assay. These quantities are studied in detail for the various standard waste matrices selected on the JRC-Ispra site. These finding are reported under a different heading.

Also topics such as detection limits and MDAs for the various standard waste matrices will be the subject of a separate report.

11.2.3 Analysis methods

The detection of neutrons in short time intervals allows the application of the so-called neutron correlation analysis, also known as neutron multiplicity counting. This method is used in passive neutron counting where the quantity of spontaneously fissioning isotopes is estimated through the determination of the spontaneous fission rate of the measured item. The spontaneous fission rate is determined, together with other sample parameters, through the measurement of neutron detection events in short time intervals (gate) using a so-called neutron multiplicity analyser. For each short time interval the number of neutrons detected is recorded and stored in a frequency distribution of signals in the gate. Each neutron detection opens a time gate for investigation of how many (subsequent) neutrons are detected during the time interval the gate is open. At the end of the measurement the total number of neutrons detected is determined simply by counting the number of gates opened during the measurement time. The advantage of this method, however, is that by forming the 1st and 2nd factorial moment of such measured frequency distributions also the number of neutron pair events and neutron triple events (observed within the length of the gate) can be determined. In this manner three independent quantities (neutron single counts, pair counts, and triple counts) are measured from the same signal pulse train. A subset of the single, double and triple counts are the correlated signal multiplets (singles, doubles and triplets) where correlated signifies neutron signals originating from the same fission event. In this way the correlated signal multiplets are each proportional to the spontaneous fission rate, allowing for the determination of the fission rate together with two other parameters related to the spontaneous fissile sample, without requiring the knowledge of the neutron counting efficiencies.

In nuclear safeguards this method is applied routinely for mass determination of plutonium in bulk samples. The method however is also applied with significant advantage for assay of plutonium in waste. In contrast to assay of bulk samples in nuclear safeguards, in waste assay where the location of the spontaneous fissile material is unknown within the waste matrix, the neutron counting efficiency, often cannot be considered known. In this case the standard method is to determine the spontaneous fission rate (proportional to the mass of spontaneous fissile material) along with the neutron counting efficiency, and a third parameter. The third parameter is often chosen to be the ratio (called the “ α ratio”) of (α , n) reaction neutrons to spontaneous fission neutrons emitted from the waste item.

The standard model, called the Point model, in neutron correlation analysis is given in the equations below. On the left hand side (R_1 , R_2 , R_3) are the measured single neutron count rates, the correlated doubles rate, and the correlated triples rate, respectively. On the right hand side are the sample and detector parameters.

Box 1. The basic Point model equations in neutron correlation analysis.

$$R_1 = F_s v_{s(1)} \varepsilon M (1 + \alpha)$$

$$R_2 = F_s v_{s(2)} \varepsilon^2 M^2 \left[1 + (M - 1)(1 + \alpha) \frac{v_{s(1)} v_{I(2)}}{(v_{I(1)} - 1) v_{s(2)}} \right]$$

$$R_3 = F_s v_{s(3)} \varepsilon^3 M^3 \left[1 + 2(M - 1) \frac{v_{s(2)} v_{I(2)}}{(v_{I(1)} - 1) v_{s(3)}} + (M - 1)(1 + \alpha) \frac{n_{s(1)}}{(v_{I(1)} - 1) v_{s(3)}} \left\{ v_{I(3)} + 2(M - 1) \frac{v_{I(2)}^2}{(v_{I(1)} - 1)} \right\} \right]$$

where the parameters are:

F_s spontaneous fission rate of sample;

ε neutron counting efficiency;

$\alpha = \frac{s_\alpha}{F_s n_{s(1)}}$ alpha ratio, i.e. ratio of (α , n) neutrons to spontaneous fission neutrons;

$M = \frac{(1-p)}{(1-pn_{I(1)})}$ the neutron self-multiplication factor;

p fast fission probability for a neutron born in the sample;
 $v_{s(x)}$ and $v_{l(x)}$ factorial moments of order x ($x = 1, 2, 3$) of the pv distributions for emission of 1, 2, 3 neutrons from either a spontaneous (s) or induced (l) fission event.

In bulk or dense samples of fissionable materials (several hundred grams) the self-multiplication factor can reach values of $M > 1.2$. In such cases the majority of the correlated double and triple neutrons originate from induced fission. In waste assay however, where the amounts of fissionable material is small or distributed in a large volume, values of M are very near 1.0. Notice in the Point model that for $M = 1$, meaning that the fast fission probability p is equal to 0 i.e. induced fission is absent, many of the terms disappear and the equations becomes much simplified. In this case neutrons are only produced from spontaneous fission and from (α, n) reactions. The standard way of applying the Point model in waste assay is to assume $M = 1$, and solve the equations for the spontaneous fission rate (proportional to the fissile mass), together with the neutron counting efficiency, ε , and α .

The limitations of applying the passive neutron correlation technique, as described above, can also be understood from the Point model equations. Obviously the model assumptions that parameters such as ε , α and M can be treated with single values (hence the name Point model) for the entire waste volume may not be exactly fulfilled in practical cases.

The design of the neutron detector head, for example, attempts to avoid large variations of ε by placing neutron detectors to the extent possible on all surfaces towards the waste item to achieve an approximate 4π counting geometry.

Also the fact that ε appears in the third power in the equations for R_3 means that it is necessary to use neutron detector heads with high neutron counting efficiencies. For this purpose many neutron detectors are required in the design. For bulk sample assay a practical requirement for multiplicity counting is roughly $\varepsilon > 0.25$ (yielding $\varepsilon^3 = 0.0156$). In waste assay a lower neutron counting efficiency can to some extend be compensated by longer measurement times.

Similarly the value of ε cannot be expected to be determined very accurately due to higher order powers with which ε appears in the equations for R_2 and R_3 . The fission rate (F_s), as the sought after quantity, on the other hand appear linearly in all three equations and will be better determined. This is the main advantage in neutron multiplicity counting when applying the factorial moments, rather than the probabilities, of the measured signal frequency distributions.

When inserting $M = 1$ in the Point Model equations above, the equations simplify to the following:

Box 2. The simplified ($M=1$) Point model equations in neutron correlation analysis.

$$R_1 = F_s v_{s(1)} \varepsilon (1 + \alpha)$$

$$R_2 = F_s v_{s(2)} \varepsilon^2$$

$$R_3 = F_s v_{s(3)} \varepsilon^3$$

where the symbols used are the same as in Box 1.

For waste measurements the solution for the three unknowns F_s , ε and α , is easily found by re-arranging the simplified equations, and using the measured rates R_1 , R_2 and R_3 :

- c) Step 1: calculate the ratio $\frac{R_1 R_3}{R_2^2}$ to yield the value of α ;
- d) Step 2: calculate (and inserting α) the ratio: $\frac{R_1 R_2}{R_3}$ to yield the value of F_s ;
- e) Step 3: calculate ε from the equation of R_1 (and inserting α and F_s).
- f)

Waste matrices with high properties for neutron slowing down and absorption, such as hydrogenous waste, have neutron counting efficiencies significantly lower than for the void matrix. This is the case for two of the waste categories at JRC, Technological Combustible and Cemented waste matrices which both have a relatively high hydrogen content.

In such cases, where the properties of the waste matrix reduce the neutron counting efficiency, the third equation (R_3) may become unusable due to high measurement uncertainty (even when long measurement times are applied). In such cases only the equations for R_1 and R_2 are usable, allowing for only two quantities to be determined i.e. the spontaneous fission rate (F_s) and a 2nd Point Model parameter, neutron counting efficiency ε . In this case the parameter α is estimated and inserted. This procedure gives the best accuracy largely because the α parameter only affects the equation for R_1 . The parameter ε on the other hand is better left as unknown because of the strong influence in the equation for R_2 .

For the waste drums with very low content of spontaneously fissile material, a situation can occur where the measured R_2 is still significantly higher (say 3 times) than the background R_2 for a given matrix type, whereas R_3 is not distinguishable from the background R_3 . In these cases the standard set of equations (above) no longer works, and further assumptions need to be made. The best way forward is to not apply the equation for R_3 and instead assume a standard value for α , and determine the two unknowns F_s and ε from the equations for R_1 and R_2 (see Box 2). In this case F_s (and the mass) is determined simply from forming the ratio $\frac{R_1^2}{R_2}$ and inserting the α parameter.

Once the spontaneous fission rate F_s is determined, the conversion to mass of ^{240}Pu effective ($^{(m)}\text{Pu}_{eq}$) is done in the standard way [22].

When the isotopic composition is considered known, either from a gamma scanning measurement, or from prior knowledge of the nuclear material, the total Pu mass is determined from $^{240}\text{Pu}_{eq}$ mass and the knowledge of abundance ratios of the Pu isotopes [22].

Annex 1 of [21] shows the template used in the passive measurements with some data from an actual measurement.

11.3 METHOD VALIDATION

The low-level radioactive waste in the JRC Ispra site, is divided into waste categories at the time of packaging. The validation of the passive/active neutron station utilises the same waste categories in form of simulated waste matrices. These simulated waste matrices used the same standard waste container (220 L drum) as for the real waste, and have in addition vertical re-entrant tubes inserted at various radii, to allow to accommodate the Pu standards within the waste matrix.

The validation procedure included placing the Pu standards at the central position (the position yielding the lowest neutron detection efficiency) for the purpose of verifying that mass determination would indeed provide results reflecting the actually applied Pu mass. The range of Pu mass was chosen to be representative for what could be expected in the waste.

In a 2nd series of measurements, a specific Pu standard was placed in various positions in the re-entrant tubes of each simulated drum matrix. The purpose was to verify the ability of the analysis method to determine the correct Pu mass, in conditions where the neutron detection efficiency would vary strongly (with sample position).

The procedure also recognised that in particular when the neutron correlation technique is applied to low spontaneous fission quantities, disturbances in form of multiple electrical noise signals in short time intervals are able to mimic the detection of spontaneous fission neutron bursts, and thus be particularly disturbing. In fact, this kind of disturbance causes the practical limitation for the application of the triple correlation method (applying the measured singlet, doublet and triplet count rates) to small quantities of spontaneously fissile material. For these reasons, special attention in the method validation was paid to demonstrating the suppression of electronic noise. Another limiting factor is due to particularly absorbing waste matrices. For the waste on the JRC Ispra site, this is the case of for waste conditioned in cement. Due to the high water content of cement, the neutron detection efficiency would reduce from 12.24% in the empty cavity to around 1% for matrices with more than 400 kg of cement. For waste matrices causing a very low neutron detection efficiency, the triplet count

rate (R_3) from small quantities of Pu is indistinguishable from the triplet background. In these cases only the singlet (R_1) and Doublet (R_2) count rates were applied in the analysis.

11.3.1 Validation of methods

The mass determination by the neutron correlation technique is often characterised as an absolute method, in contrast to a calibration method, because the mass is determined directly from measured counting rates, and some nuclear data (however with some assumptions about the Pu such as fraction of $^{240}\text{Pu}_{\text{eff}}$ of total Pu, and the alpha ratio of the PuO_2 material). The determined Pu mass was compared to the actual mass inserted by observing the mass discrepancy while taking into consideration the measurement precision as a propagation of the uncertainty of the measured counting rates.

For most drum matrices the neutron triple correlation method, determining from a set of three equations the quantities mass, neutron detection efficiency and the alpha ratio, was applied. For the cement matrix, where only neutron Singlet and Doublet count rates could be used, the two-equation system was used to solve for the Pu mass, and the efficiency while assuming the alpha ratio. However, due to the need to observe also assumptions of the neutron self-multiplication, and the mass ratio of $^{240}\text{Pu}_{\text{eff}}$ of total Pu, a similarly quality result was achieved simply by calibrating the measured Doublets rate (R_2) response against the inserted $^{240}\text{Pu}_{\text{eff}}$ mass.

In both cases, the mass determination were done for variable masses positioned in the centre position, and for a specific mass positioned at various locations within the drum matrix.

11.3.2 Performance characteristics

The accuracy of the mass determination was observed to be in the range of 10-30% for most waste matrices. For the cement matrix however even higher deviations from the inserted mass were observed.

As the relative mass accuracy was often observed to be comparable to 2-3 standard variations of the measurement uncertainty, it was concluded that longer measurement times than the standard 1800 s should have be adopted.

11.3.3 Background stability, noise level, spurious signal detection

The industrial environment of WCS on the JRC-Ispra site is prone to produce electrical noise detector systems. Charge collection pre-amplifiers as used with ^3He neutron counters in particular are sensitive to micro-currents on electrical ground lines and to noise generated from electrical motors on the same power net. Various methods for reducing electrical noise in the detection system were tried out, including RC filtering on HV supplies, termination of 50 Ohm signal transmission lines, use of low-noise transformer based power supplies.

Other measurements performed during the testing of the instrument, not reported here, includes measurement of the analogue pulse height spectrum of each electronics chain, and measurement of the plateau curve after the final setting of gain and discriminator level in each individual electronics chain.

Neutron background counting is useful for investigating effects such as electrical noise and double pulsing in the digital output due to faulty amplifiers. In the final setup the neutron background count rate of the 18 individual electronics chains is obtained as shown in Figure 30.

The background count rates show some variation among individual chains. This is partly due to variable thickness of the external neutron shield although the variations are not consistent. The variation of the individual count rates are too large to be explained by variations in ${}^3\text{He}$ pressure and must be attributed to different sensitivity to electrical noise in the individual electronics chains. Although generally speaking the modules towards the bottom of the instrument show lower count rates which is to be expected.

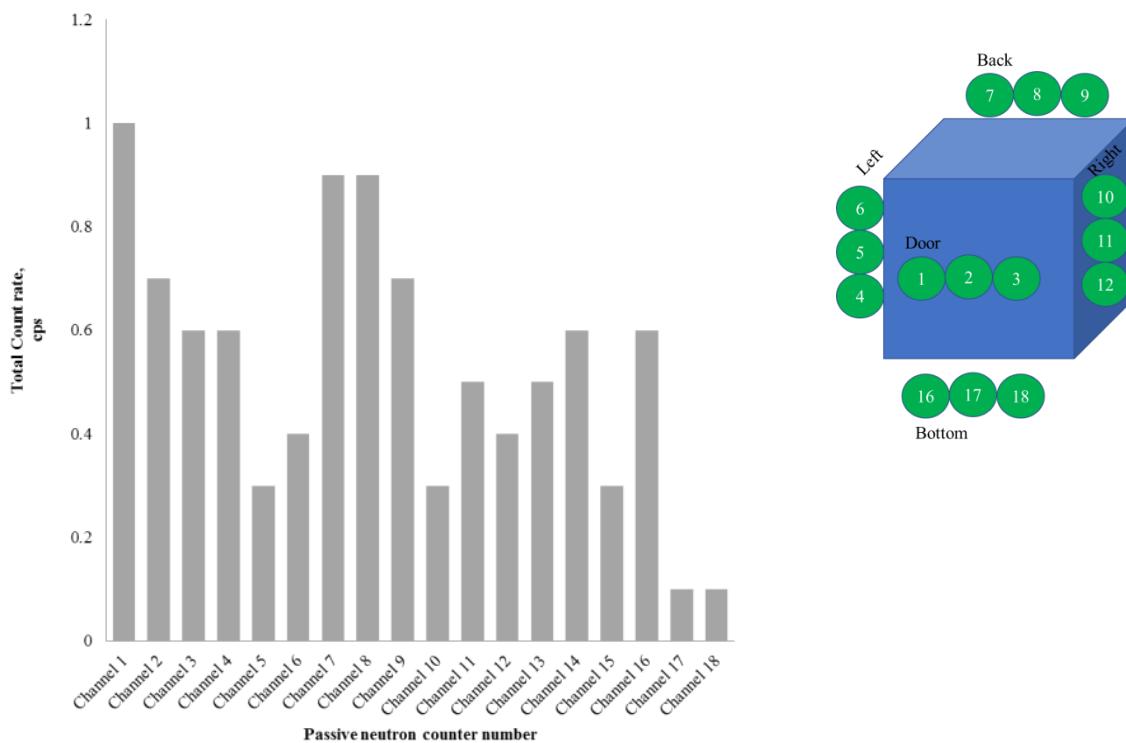


Figure 30 - Neutron background count rate measured in the 18 individual electronics channels.

An example of measured background count rates measured overnight in the passive/active neutron station is given below. The rates are obtained after hundreds of repetitions of 100 seconds duration with eliminations of repetitions with rates deviating more than four standard deviations from the mean values. The result of the measured background is (over many hours of measurement):

1. Singlets rate: 10.036 ± 0.008
 2. Doublets rate: 0.949 ± 0.003
 3. Triplets rate: 0.290 ± 0.002
- g)

where the quoted uncertainty equals one standard deviation of the measurement.

Such background values will change over time by at least 10% due to variation in the actual neutron background. Also the presence of different non-radioactive waste matrices in the drum cavity will have an impact on the background rates.

A ${}^{252}\text{Cf}$ standard source is useful for checking the performance of a neutron counting instrument. ${}^{252}\text{Cf}$ undergoes spontaneous fission similar to the even mass number Pu isotopes. The source is used regularly (daily) to check that the neutron detection system is working correctly prior to the measurement of waste drums.

The histogram in Figure 31 shows the singles count rates in each of the 18 counting chains measured with a ${}^{252}\text{Cf}$ neutron source placed in the centre of the otherwise empty.

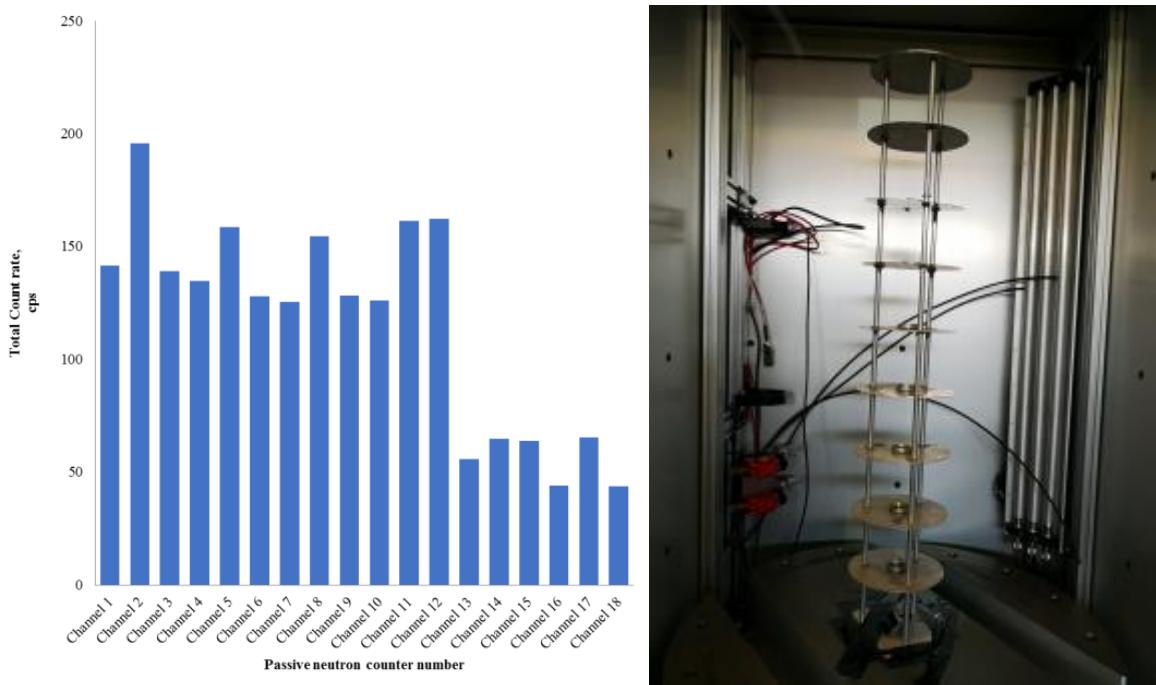


Figure 31 - Measurement of the neutron signal count rate in the 18 individual channels, ^{252}Cf standard source in central position.

The neutron singles count rate variations based on a central ^{252}Cf source can largely be understood from geometrical considerations alone:

1. Of the six sides surrounding the drum cavity, the four vertical sides show higher count rates than the top and bottom modules. This is to be expected as these ^3He tubes are longer than the one top and bottom, and the surface area of the vertical sides is larger than the top and bottom.
2. On each side the ^3He tubes in the centre module (three modules on each side) show higher count rate than the peripheral ones. This is to be expected as the central modules have more polyethylene in the vicinity than the peripheral modules, taking into account that all module surfaces are covered in a 1 mm Cd liner, but the surface between modules is not.

h)

i)

11.3.4 Quality control tests, diagnostics of the acquisition system

As mentioned above the neutron correlation analysis was performed using a standard Canberra JSR-14 signal analyser. However the introduction of the TDS32 with the primary task of merging the 18 signal lines into one, also allowed for diagnostics on the signal pulse train. This was done by off-line analysis of the list-mode data transmitted to the host PC over the USB3.0 connection.

Large neutron detection systems with many parallel signal processing chains are known to be prone to electrical disturbances and noise. When in addition the detection system is operated in an industrial environment often with electric motor drives in operation, special care must be observed to avoid degradation of the measurement data due to electrical noise. A useful remedy is to divide the total measurement time into multiple repetitions of smaller measurements times, and to discard outlier repetitions, caused by electric noise, through outlier criteria such as setting a maximum deviation from the mean to e.g. three sigma of the measurement uncertainty. In practice, when other activities were ongoing during the measurements, as many as 30% of 100 s measurement could be rejected by the outlier criterion. In these cases the total measurement time was extended until the “good” repetitions represented a total measurement time of at least 30 min.

To overcome the problem of imperfection of the applier circuits producing double signals in the digital signal pulse train, the artificial dead-time of 1 μs was introduced in the signal mixing operation of the TDS32. Note that this remedy is only acceptable because the count rates (of singlets, doublets and triplets) are low in waste assay. Had the count rates been higher (above 1000 s^{-1}), a significant dead-time effect would influence the measured data.

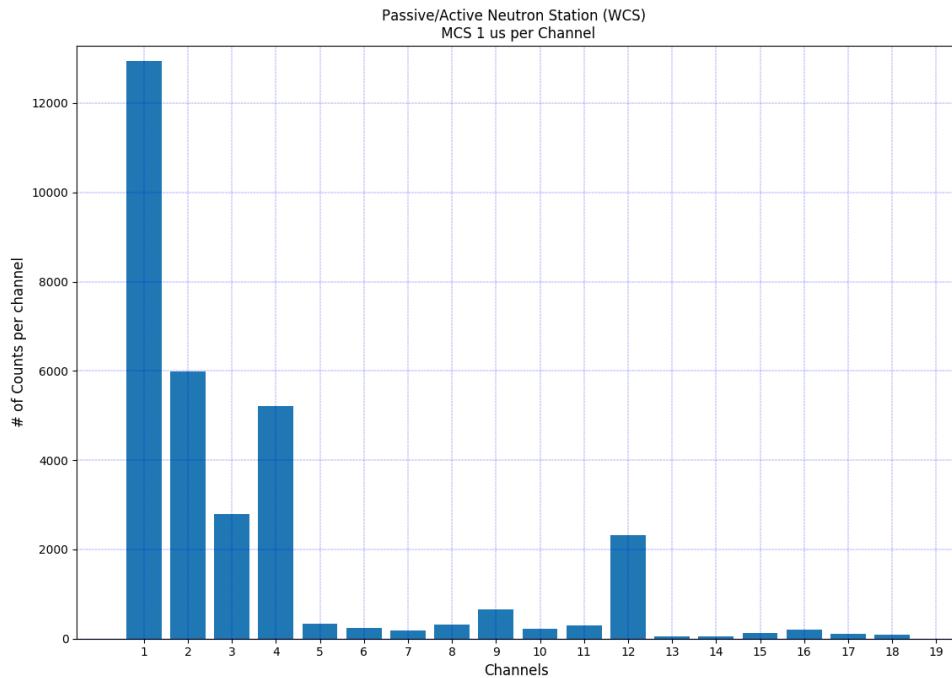


Figure 32 - Plot of counts in each of the 18 fission neutron counters during a background measurements prior to inserting the 1 μs artificial dead-time in the pulse processing.

The double pulsing problem observed in the passive/active neutron station was investigated in detail prior to starting the characterisation of waste items. By means of lists of recorded time-stamps, produced by the TDS32, the time intervals between subsequent pulses were analysed. Figure 32 shows the number of counts in each of the eighteen signal channels for a long background measurement. This is prior to the introduction of the dead-time of 1 μs in the pulse processing. The figure shows abnormally high counting rates in the channels 1 to 4, and 12 of the fission neutron counters. This difference in counting rates is clearly caused by false double pulsing in these individual counting chains.

The double pulsing effect was investigated in further detail. In Figure 33 the number of events recorded where a 2nd pulse follows within an interval of 1 μs of any given pulse, is shown, and in addition the channel number is noted both of the initial (triggering) pulse, and the subsequent pulse. The example given here is from a very long background measurement of several days.

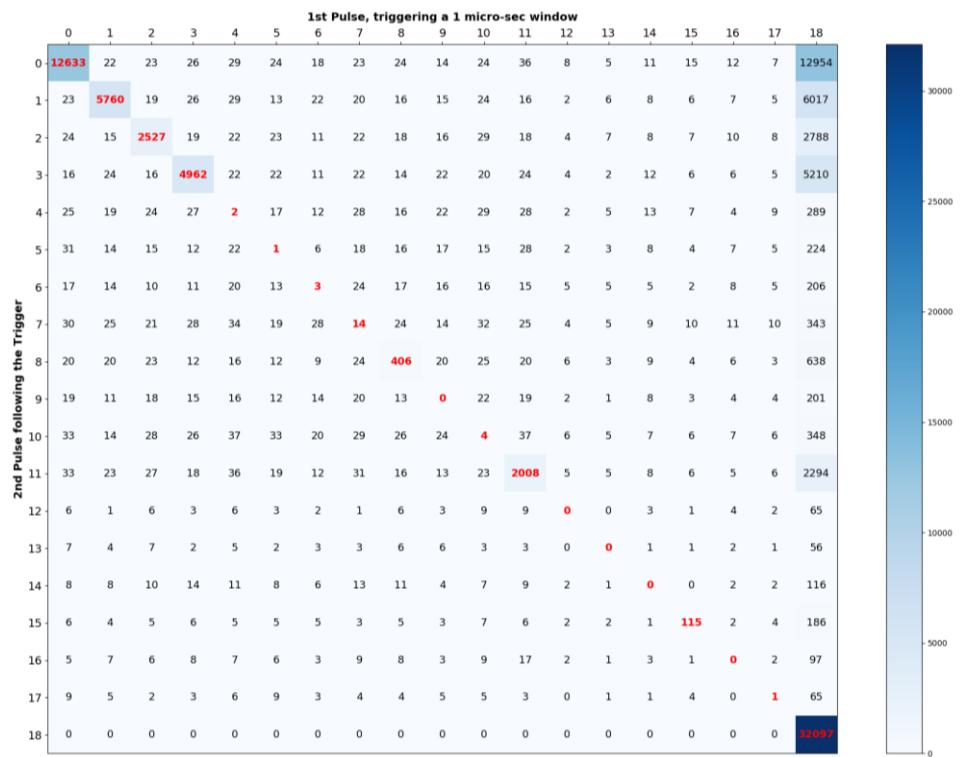


Figure 33 - Events of two subsequent pulses given as channel number of 1st pulse and channel number of 2nd pulse on the condition that the 2nd pulse falls within 1 μ s of the 1st pulse.

Figure 33 shows the problem of double pulsing as observed in the passive/active neutron station. At the low background count rates, singlets around 10 s^{-1} , the high number of events with two pulses within a 1 μs period should not occur, and can certainly be attributed to false pulse in the detection system. In particular, the events on the diagonal (channel 0 to 3 and 11, in red colour) are particularly frequent. This was also observed in Figure 33. The diagonal indicates double events in the same amplifier within 1 μs . These events are clearly artificial, and due to malfunction of the JAB-01 amplifier circuits. The off-diagonal counts are caused by a TTL digital signal produced on one single chain inducing charge into the pre-amp on another chain causing a false signal.

Figure 34 shows the same signal pulse train, and 1st and 2nd pulse-analysis as in Figure 33 but with the 1 μs filter activated. The electrical noise has clearly been removed, and the operation of the filtering is working correctly.

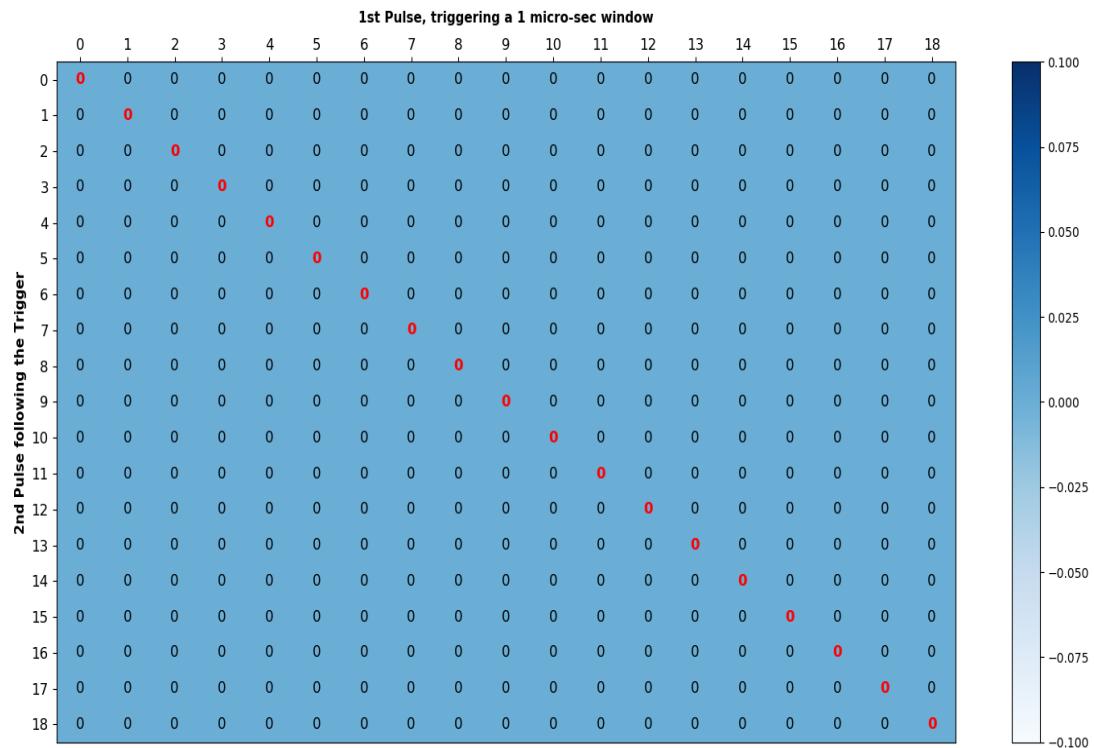


Figure 34 - Events of two subsequent pulses given as channel number of 1st pulse and channel number of 2nd pulse (same as Figure 33 but with 1 μ s filter following the 1st pulse).

For the neutron correlation analysis, the measured Singlets (total counts), Doublets and Triplets are the important measured quantities. Figure 32 shows the examples of background measurements in the empty drum cavity with and without the 1 μ s filtration inserted in the TDS32 device. After filtration the singles count rate reduced by roughly 12%. The change for doublets and triplets count rates was much more significant being 26% and 53%, respectively. Clearly the analysis of small quantities of spontaneous fissile material would be impossible without the filter inserted. Given the significant effect of the filtering on the measured signal triplets, the neutron correlation analysis as proposed in [21] simply would fail without the application of filtering. In addition, by significantly reducing the general noise level, the filtering also improves the achievable detection limit (MDA).

Table 18 and Table 19, show a comparison of the measured doublets and triplets count rates from background with and without signal filtration.

Table 17 - Comparison of long background measurements in empty cavity with/without 1 μ s filter (TDS32).

| Measurement description | Count time, s | Singles R ₁ , s^{-1} | Doublets R ₂ , s^{-1} | Triples R ₃ , s^{-1} |
|---|---------------|-----------------------------------|------------------------------------|-----------------------------------|
| Background without TDS32 filter, B | 71800 | 9.72 ± 0.013 | 0.941 ± 0.005 | 0.334 ± 0.005 |
| Background with TDS32 filter, B _{TDS32} | 76600 | 8.69 ± 0.012 | 0.745 ± 0.004 | 0.156 ± 0.003 |

Table 18 - Variation of doubles after filtration for background measurements, in %.

| Filling of standard calibration drum | Background doubles without signal filtration, s^{-1} | Background doubles with 1us filtration (TDS32), s^{-1} | Variation of doubles after filtration, % |
|--------------------------------------|--|--|--|
| Empty cavity | 0.95 ± 0.003 | 0.745 ± 0.004 | 20% |
| Air drum | 1.04 ± 0.007 | 0.904 ± 0.005 | 13% |
| MTL drum | 1.06 ± 0.006 | 1.092 ± 0.006 | -3% |
| TNC drum | 0.97 ± 0.003 | 0.939 ± 0.022 | 3% |

Table 19 - Difference in triplets after filtration for background measurements, in %.

| Filling of standard calibration drum | Background Triples without signal filtration, s^{-1} | Background Triples with 1us filtration (TDS32), s^{-1} | Variation of triples after filtration, % |
|--------------------------------------|--|--|--|
| Empty cavity | 0.29 ± 0.002 | 0.156 ± 0.003 | 46% |
| Air drum | 0.35 ± 0.007 | 0.239 ± 0.005 | 31% |
| MTL drum | 0.37 ± 0.007 | 0.295 ± 0.005 | 20% |
| TNC drum | 0.32 ± 0.003 | 0.213 ± 0.015 | 33% |

The variations of the count rate are significant for the triples and doubles for background measurements of standard calibration drums with and without 1 μ s filtration (TDS32). The fact that the triplets quantity is most perturbed by electrical noise is perfectly understandable given that this quantity is derived from the 2nd factorial moment of the measured signal frequency distribution.

11.3.5 Impact from choice of HV on gas proportional counters

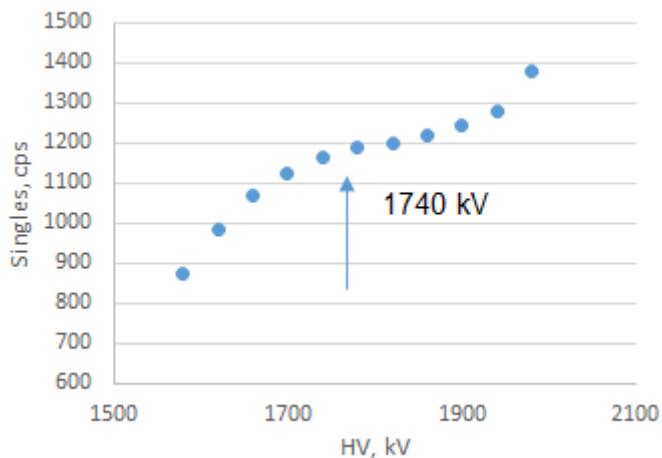
The high voltage applied to the anode wire, when within a certain range, works as a coarse gain for the analogue signal produced in the amplifier through variation of the gas-multiplication and with that the analogue signal height as function of HV setting. For this reason the HV setting can to some extend be used to reduce the double pulsing of TTL signals. For this reason the impact of HV applied to ${}^3\text{He}$ tubes was investigated.

11.3.6 Plateau

Plateau measurements, in form of signal count rate as function of HV setting, were performed using a sealed standard ${}^{252}\text{Cf}$ source. The ${}^{252}\text{Cf}$ source was placed in the standard "Air" simulation drum in the central position. The signal count rate was acquired with the 1 μ s filtration inserted.

Table 20 - Singlets, Doublets, Triplets measurements at different anode voltages.

| HV, kV | Singlets R ₁ , s ⁻¹ | Doublets R ₂ , s ⁻¹ | Triples R ₃ , s ⁻¹ |
|--------|--|--|---|
| 1580 | 873 | 87 | 5 |
| 1620 | 984 | 111 | 7 |
| 1660 | 1067 | 130 | 9 |
| 1700 | 1126 | 146 | 11 |
| 1740 | 1164 | 155 | 12 |
| 1780 | 1188 | 162 | 14 |
| 1820 | 1201 | 165 | 15 |
| 1860 | 1218 | 170 | 17 |
| 1900 | 1245 | 175 | 20 |
| 1940 | 1279 | 179 | 23 |
| 1980 | 1377 | 184 | 25 |

**Figure 35 - Plateau curve based on singlets count rate.**

The high voltage setting of 1740 V was found to be the best working high voltage of the fission neutron counting system both with respect to digital signal noise reduction and gas proportional counter operation (analogue signal height and discriminator setting), as it is shown in Table 20 and Figure 35. All further measurements were performed at this high voltage setting.

11.3.7 Background disturbances from external sources and electrical noise

Measurement of the neutron background was performed under various conditions to investigate the external influences. These investigations were all performed with the 1 μ s filtration inserted. Although electrical motor drives had been found to disturb measurements at other occasions, the measurement in Table 21 where the crane was deliberately operated simultaneously did not appear to be disturbed. Also the presence of the neutron sources in the nearby safe, did not seem to influence the neutron background measurement. For this reason the influence of these two kinds of external disturbances remained inconclusive.

Table 21 - Comparison of the background measurement of empty cavity parameter measurement time with 1 μ s filter (TDS32).

| Measurement description | Count time, s | Singlets R ₁ , s ⁻¹ | Doublets R ₂ , s ⁻¹ | Triplets R ₃ , s ⁻¹ |
|---|---------------|---|---|---|
| Background (Artefact no PuO ₂ in safe) | 76600 | 8.69 ± 0.012 | 0.745 ± 0.004 | 0.156 ± 0.003 |
| Background (Artefact movement of crane) | 800 | 9.576 ± 0.122 | 0.849 ± 0.04 | 0.206 ± 0.025 |
| Background | 1000 | 10.63 ± 0.114 | 0.82 ± 0.035 | 0.247 ± 0.043 |
| Background | 13200 | 10.883 ± 0.032 | 1.003 ± 0.011 | 0.264 ± 0.009 |

11.3.8 Blank waste matrices

The neutron background counting rates in the fission neutron counters depends on the type of in-active matrix material (blank material) is loaded into the drum cavity. This count rate is called “blank” count rate in the following. Strongly absorbing matrices (e.g. high hydrogen content) can reduce the measured neutron blank rates (compared to an empty cavity), whereas matrices with high atomic number materials, or high density materials, can cause increased neutron blank rates through interactions of atmospheric muons. It is well-known that muon interactions with such matrices can cause emission of bursts of spallation neutrons. For this reason it is expected that the neutron blank rates varies as function of the matrix material in the drum. In order to subtract the correct signal blank, a blank measurement with of the relevant non-active matrix material should always be performed immediately before, or after, measuring actual waste of a certain matrix type.

The measured blank count rates as function of matrix material is shown in Table 22 below.

Table 22 - Comparison of blank measurements of standard calibration drums with 1 μ s filtration (TDS32).

| Matrix | Meas time [s] | Measurement raw data | | | | | |
|--------|------------------|----------------------|-------------------|--------------------|-------------------|--------------------|-------------------|
| | | Singlets rate | | Doublets rate | | Triplets rate | |
| | | [s ⁻¹] | 1 σ [s] | [s ⁻¹] | 1 σ [s] | [s ⁻¹] | 1 σ [s] |
| Air | 178500 | 9.662 | 0.008 | 0.906 | 0.003 | 0.255 | 0.003 |
| MTL | 705900 | 10.664 | 0.004 | 1.089 | 0.002 | 0.327 | 0.002 |
| TNC | 181800 | 9.556 | 0.008 | 0.876 | 0.003 | 0.224 | 0.002 |
| TC | 54800 | 9.251 | 0.015 | 0.853 | 0.005 | 0.248 | 0.005 |
| CMT | 54100 | 9.149 | 0.015 | 0.816 | 0.005 | 0.227 | 0.004 |

Table 22 shows examples of long blank measurements with different non-active matrices inserted. An increase of the count rates as function of the amount of metal in the matrix and drum. As expected, the triplets count rate is affected more than doublets and singlet count rates. This is because spallation neutrons often are emitted in high multiplicity bursts which produce relatively more triplets than doublets and singlets, respectively.

In addition, the component of the neutron blank originating from interactions with high energy particles may vary with atmospheric conditions. This is an additional reason for always performing a blank measurement before/after drum assay.

Angular dependence of detectors response

The passive/active neutron station has a base plinth in the cavity for supporting the waste drum. The manufacturer of the station made it possible to rotate the base plinth during measurement. The purpose of this is to eliminate variations in neutron detection efficiency

(passive and active measurement) and neutron interrogation (active measurement) due to the geometrical limitations of the instrument (i.e. a parallelepiped shaped drum cavity, neutron generator located in one fixed position). However if rotation would not be required to compensate these effects, some advantage could be gained in terms of electrical noise reduction by not operating the electrical motor drive.

Measurements were carried out with the ^{252}Cf standard source placed in the centre (vertical) plane at the periphery of an empty 220 L drum. The drum was rotated in steps of 15° and the total count rate was measured for at least 20 min in each position. The purpose was to investigate the angular dependence of the combined signal rate in the 18 detector modules. The neutron count rate was recorded in all of the 24 source positions. The result of these measurements is showed in the Figure 36.

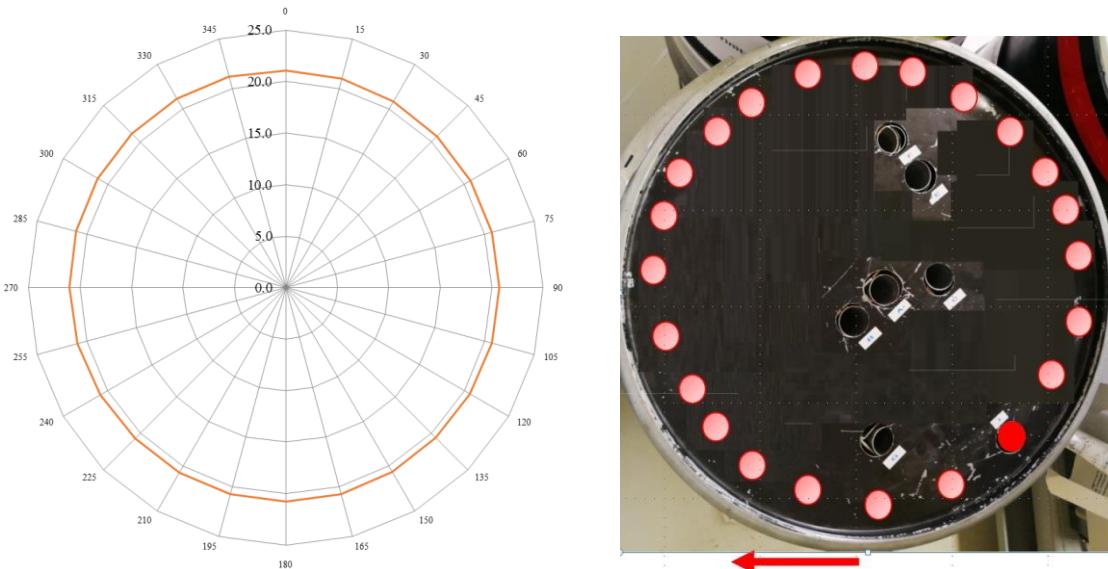


Figure 36 - Angular dependence of detectors response

In the passive neutron correlation analysis (see e.g. ref 2 of [21]) the knowledge of the die-away time of the neutron detection system is needed. Also in the active mode of operation, the DDA analysis requires a much shorter neutron life-time of in the detector modules (cadmium covered polyethylene slabs) than in the moderator for the generator source neutrons (the graphite mantle). A well-defined die-away time, a single exponential decay, is generally achieved by reducing the width of the polyethylene moderator (under-moderated) to less than the optimal thickness if only detection efficiency would be optimised, and by covering all module surfaces in a cadmium liner.

11.3.9 Measurements of known PuO_2 samples

Sealed sources of PuO_2 materials were inserted in the re-entrant tubes of the simulated 220 L waste drums. The samples used are described in (§8.2.2). The isotopic composition has been updated to approximately the day of measurement, and the masses of $^{240}\text{Pu}_{\text{eff}}$ and total Pu is given in Table 8. In each measurement the JSR-14 neutron correlation counter delivers the measured totals count rate (singlets), the rate of correlated doublets, and the rate of correlated triplets. In [21] these quantities are referred to as R_1 , R_2 and R_3 , respectively. From the determination of the spontaneous fission rate F_s [21], the mass of spontaneous fissile material, $^{240}\text{Pu}_{\text{eff}}$, is determined simply from nuclear data of ^{240}Pu . The mass of $^{240}\text{Pu}_{\text{eff}}$ is given by:

$$\text{Mass}(\text{Pu}_{\text{eff}}) = \frac{F_s}{473.5 F_{\text{iss}}/\text{g s}}, \quad (24)$$

where F_s is the calculated spontaneous fission rate of the measured sample and the number of spontaneous fissions per gram per second of ^{240}Pu is $473.5 \text{ s}^{-1}\text{g}^{-1}$.

From the knowledge of the isotopic abundance of the Pu sample at the time of measurement, the total Pu mass is determined using the formula (1) in Chapter 10.1.2.

The annexes 1 -5 of [20] are dedicated to each of the types of investigated waste matrices (except for cement). Notably these are the waste matrices of the waste currently being produced on the JRC-Ispra site. The matrices are:

1. AIR drum, for drums of light matrices (with minimum impact on neutron transport)
2. MET drum, waste of different types of metal
3. TNC drum, technological waste of non-combustible type
4. TC drum , technological waste of combustible type (often hydrogenous waste)
5. Cement drum, representing waste conditioned in cement

Each Annex has two tables. One table shows the measurement of Sample 5 in different locations of the drum to verify to which degree the mass is determined correctly at different sample locations. The re-entrant tubes nominated A, B, C, and D, are radial locations in the drum, where A is the centre position and D near the drum periphery. The numbers 1-4 represent different heights in the re-entrant tube starting with position O on top, and position 2 is the middle vertical position. In the tables the measurement time, and the measured rates of singlets, doublets and triplets (R_1 , R_2 and R_3), together with their uncertainties, are given as the measurement data.

Through the neutron correlation analysis, by solving a system of three equations, the mass has been determined together with the two sample parameters: neutron counting efficiency (ε), and the alpha ratio (α). The uncertainties on these quantities given in the tables represent one standard deviation as propagated from the uncertainties of the measured counting rates. For convenience, the mass uncertainty is given here as relative to the mass value in percent. The assay accuracy of the measurements is presented in the tables of the Annexes [20], as a column called “ Δmass ”. This quantity is calculated as:

$$\Delta\text{mass} = \frac{\text{mass}_{\text{declared}} - \text{mass}_{\text{measured}}}{\text{mass}_{\text{declared}}} \cdot 100, \quad (25)$$

or, in other words, the percent deviation from declared mass. The accuracy of the assay result (Δmass), should be viewed in connection with the measurement uncertainty given in the table. In most measurements, the assay result does indeed fall within 1 to 2 times the standard deviation of the measurement (as should be expected). In hindsight, the measurement time should arguably have been chosen longer than the usual $\frac{1}{2}$ hour in order to improve the uncertainty of the measurement result.

Clearly, the neutron counting efficiency varies as function of the sample position due to the different amount of neutron moderator and absorber surrounding the sample in each location. The determination of the two sample parameters, ε and α , is not expected to be very accurately due to the underlying equations (see [21]). The efficiency appears in the equations in higher powers, and the alpha ratio only in one of the three equations. The spontaneous fission rate (proportional to the mass), however, appears linearly in all three equations, and is thus determined more accurately (this is in fact the strength of the correlation analysis method).

For the purpose of checking linearity of the mass assay as function of fissile mass, samples of different mass were measured in the centre position of each matrix type (position A2). The measurement results are shown in the 2nd table of the annexes 1–4 in [20]. The analysis results are again given as Pu mass, alpha ratio and neutron counting efficiency, determined in the same way as the 1st table. Also in this table, the assay result in most cases fall within 1-2 standard deviations of the measurement uncertainty.

Annex 5 in presents the case of the cemented drum. Unfortunately only few measurements were done for this matrix. The high content of hydrogen, bound in water, in the cement causes moderation of the neutron energy through scattering processes. In addition a fraction of the spontaneous fission neutrons are absorbed in hydrogen rather than being emitted from the drum. Due to these effects the neutron detection efficiency for the cemented drum is much lower than for the other matrices. This is seen clearly in the tables of Annex 5 in [20] where the measured values of R_3 are not significantly above the level of the blank measurements. In these cases the neutron correlation analysis using R_1 , R_2 and R_3 will not work.

11.3.10 Analysis of measurements of Pu in the cement matrix

In the case of the cement matrix the standard neutron correlation analysis cannot be applied due to the low neutron detection efficiency. As the measured triples rate (R_3) is proportional to the neutron detection efficiency in the third power, this measured quantity suffers most from the low efficiency. As suggested in [21] in such cases the mass can be determined using the equations for R_1 and R_2 while assuming the value of alpha (α) value (the ratio of (α , n) reaction rate to spontaneous fission neutron emission rate), thus determining the spontaneous fission rate (proportional to the mass) together with the neutron detection efficiency.

This procedure was applied to achieve the results in Annex 5 in [20]. The mass, calculated from R_1 and R_2 , deviates in the range 16-20% from the real value. In these calculations the correct α value (calculated from the known isotopic composition of the samples) were inserted (see values in Annex 5 of [20]). In real cases the value of α would need to be estimated, contributing a substantial uncertainty to the mass calculation. Notice also that the calculation of the neutron detection efficiency returned a value around 1-2%. This is due to the strong neutron absorption of very hydrogenous matrix.

In many practical cases, however, this approach is prone to large uncertainty due to the difficulty in estimating the α value. Particularly in waste suspected of containing multiple α -emitters, e.g. waste containing minor actinides, a wrong estimate of α can impact negatively on the determination of the spontaneous fission rate applying the two-equation method [21].

In this case, it may be preferable to avoid the large uncertainty due to the unknown α value, and accept the uncertainty due to the unknown detection efficiency, and determine the mass of $^{240}\text{Pu}_{\text{eff}}$ (proportional to the spontaneous fission rate) only from the measured value of R_2 . For this purpose a calibration of known $^{240}\text{Pu}_{\text{eff}}$ masses against the measured R_2 is required for the given matrix type. The large uncertainty in the $^{240}\text{Pu}_{\text{eff}}$ mass determination is mainly due to the unknown detection efficiency in the actual drum.

Notice that avoiding the contribution of uncertainty of α (as in the equation for R_1) to the mass estimate, is only achieved when the assumption that the (α , n) reaction neutrons do not cause additional induced fission events ($M = 1.0$) is true i.e. for small masses of Pu, or Pu diluted (distributed) in the waste matrix.

The Doubles rate (R_2), after subtraction of the blank value, against the known mass of $^{240}\text{Pu}_{\text{eff}}$ for the cemented drum for the cement matrix is given in Figure 37 below based on the data of Annex 5 of [20]. Unfortunately only three measurements were done for this simulated drum. In addition these measurements are all in the centre position (A2). More measurements are needed with different Pu masses and at different locations in the drum.

A 2nd order polynomial, intercepting at the origin, are fitted to the data as:

$$R_2 = a ({}^{240}\text{Pu}_{\text{eff}}) + b ({}^{240}\text{Pu}_{\text{eff}})^2 \quad (26)$$

where the fitting parameters are:

$$a = -0.06897 \pm 0.03863$$

$$b = 0.09401 \pm 0.01468$$

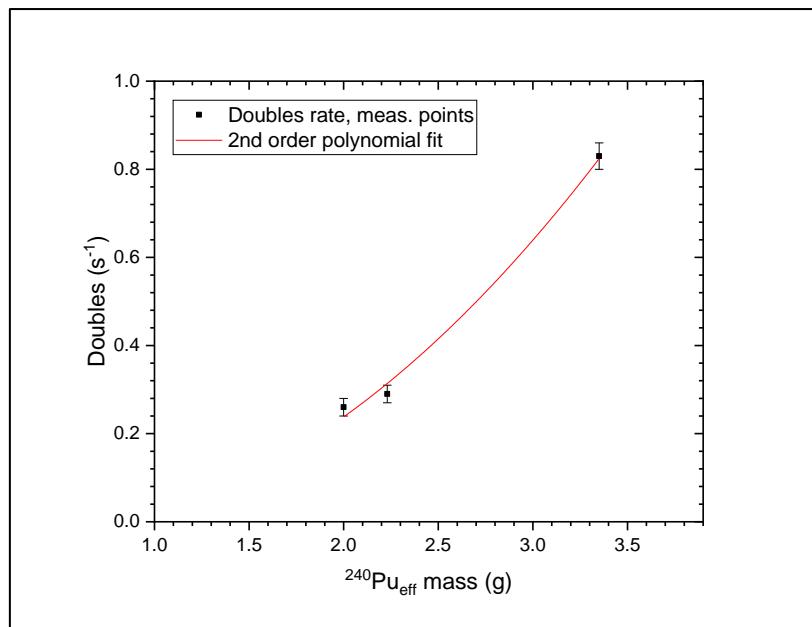


Figure 37 - Calibration of Doubles rate against $^{240}\text{Pu}_{\text{eff}}$ mass for the cement drum.

In principle a linear fit is expected according to the equation for R_2 . But given the few data points a 2nd order has been applied. The shown uncertainties are due to the measured R_2 alone. The real uncertainty would have more significant components from the unknown sample position inside the drum, and the unknown isotopic vector of Pu needed for the translation from $^{240}\text{Pu}_{\text{eff}}$ mass to total Pu mass.

To demonstrate the principle of the linear relationship between the measured R_2 and the $^{240}\text{Pu}_{\text{eff}}$ mass, Figure 38 shows the measured Doubles rate (after blank subtraction) as function of $^{240}\text{Pu}_{\text{eff}}$ mass in the case of the metal drum where more data were available.

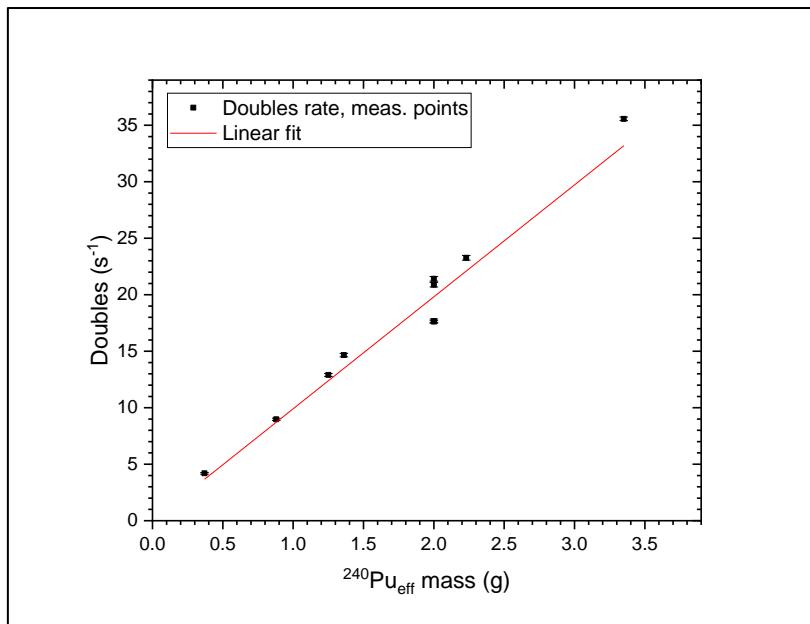


Figure 38 - Calibration of Doubles rate against $^{240}\text{Pu}_{\text{eff}}$ mass for the metal drum.

The measured R_2 values were fitted against the $^{240}\text{Pu}_{\text{eff}}$ mass. The linear fit, intercepting at the origin, is:

$$R_2 = a ({}^{240}\text{Pu}_{\text{eff}}) \quad (27)$$

where the fitting parameter is: $a = 9.90844 \pm 0.28706$.

In Figure 38, the R_2 values corresponding to the mass value 2.0 g are the measurements of Pu Sample5 (see Table 8) at different locations inside the metal drum (See Annex 2 in [20]). These values shows a discrepancy of up to 12% from the fitted value at 2.0 g solely due to the variation of neutron detection efficiency in different locations of the metal drum. For the waste matrices with stronger moderation and absorption (than the metal drum) this variation increases.

11.4 MEASUREMENT CAMPAIGN of real waste

As observed in the campaign on simulated waste drums, the blank measurement for the background subtraction in the measured signal multiples (R_1 , R_2 and R_3) to achieve the net counting rates from the waste, must be of the same type of matrix as the waste (due to the atmospheric interactions creating neutrons in the waste matrix material). In addition, the blank measurement should always be done at a time not too distant from the waste measurement (to cope with neutron background variations with atmospheric conditions). In the measurement of the real waste drums [24], care was taken to always perform the blank measurement, of the corresponding matrix material, at the most some hours prior to the waste measurement. Most often the blank measurement was performed overnight prior to the waste measurements.

All real waste drums were measured three to four times. These measurements were typically separated by several days. Thus any repetition cycle included a separate blank measurement, as well as the drum movements and positioning in the drum cavity.

At the time of the present project, a total of 12 real waste drums of the types: technological non-combustible (TNC), technological combustible (TC), and metal (MTL) were available for measurement. In none of the 12 waste drums, a Pu content above the MDA value was not observed. In spite of the careful blank measurement prior to each waste assay (see point

above), and long measurement times (of at least 1½ hours), the measured net counting rates (R_1 , R_2 and R_3) were generally near zero but negative. Only for the TC drum type, the R_1 value was near zero but consistently positive for two drums.

This fact suggests that the simulated drum matrices did not correctly represent the real waste matrices. The systematically negative net counting rates in the real waste drums indicate that they had more neutron absorbing materials (e.g. plastic or aquatic/wet materials) than the simulated non-active waste matrices. This was however not observed for the TC type drums. The minimum detectable mass (or activity), MDA, was calculated using the standard method applied in neutron coincidence counting [25]. The calibration factor, translating from the measured R_2 to the mass of $^{240}\text{Pu}_{\text{eff}}$ were calculated as linear fits to the measured quantities [24]. The minimum detectable mass (MDA) for the five matrices is given in the table below.

Table 23 - Calculated MDA values for different matrix types in the passive/active neutron station.

| Matrix | Measurement data | | | | | Calculated data | | MDA per gram of waste |
|--------|------------------|-------------------------|--------------------------------|-------------------------|--------------------------------|--|--|-----------------------|
| | Meas time | Singlets rate (R_1) | | Doublets rate (R_2) | | Calibration R_2/mass ($^{240}\text{Pu}_{\text{eff}}$) | MDA Mass of $^{240}\text{Pu}_{\text{eff}}$ | |
| | [s] | [s^{-1}] | 1σ [s^{-1}] | [s^{-1}] | 1σ [s^{-1}] | [s^{-1}/g] | [mg] | [$\mu\text{g/g}$] |
| Air | 178500 | 9.662 | 0.008 | 0.906 | 0.003 | 10.86 | 9.77 | 0.189 |
| MTL | 705900 | 10.664 | 0.004 | 1.089 | 0.002 | 9.908 | 11.7 | 0.046 |
| TNC | 181800 | 9.556 | 0.008 | 0.876 | 0.003 | 9.926 | 10.5 | 0.061 |
| TC | 54800 | 9.251 | 0.015 | 0.853 | 0.005 | 5.021 | 20.5 | 0.224 |
| CMT | 54100 | 9.149 | 0.015 | 0.816 | 0.005 | 0.1721 | 586 | 1.250 |

12 LESSONS LEARNED

12.1 GAMMA STATION (TSGS)

An initial assessment of the WCS in TGS mode is performed and some technical is solved in the period of several months. Those were the following:

1. Index PC=0 and D-Spec=0. This error happened when the D-Spec Plus was not connected to the HPGe Detector. Indeed, the TGS scan was stopped during the forward emission scan of the first layer; the measurements did not continue. When the D-Spec Plus is correctly connected to the HPGe detector, the index PC and D-Spec should increase up to 74 for PC and 75 for each scan (emission/transmission).
2. The cooling fan of the D-Spec plus did not work anymore and it was the reason why the D-Spec plus became too hot and Dead-time reached 99%. The cooling fan was substituted.
3. JOB ERROR 12 appeared on the screen when the job file performed the command SET_PRESET. The error appeared because the internal battery of D-Spec Plus did not work anymore. This error prevents to perform TGS measurements. In this case, the straight-through and dead-time correction measurements are not performed. The internal battery of the D-Spec was replaced.
4. The assay types available on *MasterAnalysis* did not give the activities of the sources inserted on the calibration drum, probably due to a wrong calibration (i.e. wrong ROIs channels). A new assay type was created and calibrated.
5. The image reconstruction of the drum for the emission energy below 100keV is not calculated and a message error appeared on *MasterAnalysis*.
6. MASTERANALYSIS ERROR 9 (in SGS mode): the program does not let the user to insert the container and master attenuation coefficients. Possible cause due to low counts of the transmission source with a MTL drum.

Some hardware limitation:

7. During the acquisition, the drum is continuously rotated and translated. Hence, it is preferable that there is no dead-time between two subsequent acquisitions. Grab times may be quite short and for this purpose the Repetitive Acquire Mode of the ORTEC® DSPEC Plus (1999) gamma-ray spectrometer is used (from the DSPEC plus-brochure.pdf) in the *MasterScan* software. The DSPEC Plus has two memory buffers: one is used to store the spectrum being acquired, whereas the second buffer can be read via Ethernet with IEEE 802.3 10 BASE2 (thin coax), which has a low speed of 10Mbits/s. The more recent Ortec DSPEC-50 does not provide the “Repetitive Acquire Mode”. Its 100BASE-T Ethernet port allows 10 times faster speeds than the DSPEC Plus, and the Repetitive Acquire Mode is replaced by the more versatile “List Mode⁸” acquisition mode. The *MasterScan* software is not compatible with it.

Some mechanical design limitation:

8. The use of a tungsten collimator in front of the HPGe detector, lower the detection limit of the important radionuclides vector, ²⁴¹Am. This is due to the interference of the mean peak of ²⁴¹Am (59.54keV) with the X-ray of tungsten of the collimator ($K_{\alpha 1}$: 59.32keV and by less extent by $K_{\alpha 2}$: 57.98keV). This X-fluorescence of tungsten is induced by any gamma sources present in the waste and by the ¹⁵²Eu source used for the determination of the gamma self-absorption in the analyzed waste drums. Due to this energy interference, analysis of ²⁴¹Am is challenging and requires very high energy resolution HPGe detector with a sophisticated program for the deconvolution in that interference energy range of the analysed gamma-ray spectra.
9. Due to the low activity of the ¹⁵²Eu transmission source (i.e. ~2.29MBq), the TSGS has a low counting rate thus is not able to perform measurement in a high density matrix (e.g. ≥CMT matrix).
10. At the same due to the high activity of the ¹⁵²Eu transmission source, for lower energies radionuclides (e.g. ²⁴¹Am at 59.7keV and 133Ba at 81keV gamma lines), the shielding material of the shutter is insufficient to reduce the background produced by it, and only TCO, TNC, and RBL matrix were analysed for the measurement campaign.
11. Due to the low activity of the available ²⁴¹Am point source (e.g. ~17MBq) and the low intensity of the ¹⁵²Eu transmission source only the TCO drum could be investigated for the ²⁴¹Am nuclear vector.

Some software limitation:

12. For the real waste with fissile materials, we do not have a quantitative evaluation (e.g. mass determination) on the activity determined by the *MasterAnalysis* but only a qualitative results can be taken (presence or not of fissile materials).
13. It is important that in order to have a correct determination of the unknown activity in a real drum, the calibration of the specific matrix should be performed using multiple sources in order to span the all energy ranges of radionuclides of interest and as well as the activity range (e.g. VLLW, LLW and ILW).

12.2 PASSIVE NEUTRON STATION

The ability to assay small quantities of Pu in a waste matrix is inevitably linked to the variation of the neutron background. It is well-known that the neutron background changes with atmospheric conditions. This is because a fraction of the neutron background is produced by cosmic ray particles and atmospheric muons. As the atmospheric conditions changes so does the neutron background count rate in the measurement device. In addition, a large waste matrix can influence the neutron count rate by altering the atmospheric background. A large hydrogenous matrix may absorb neutrons and thus produce a smaller neutron background than an empty drum cavity. In contrast, a matrix composed of metal may

⁸ In the “list mode” operation, data are streamed directly to the computer, event by event with no associated “dead period” (accuracy of 200ns).

increase the neutron background through spallation of atmospheric muons producing burst of neutrons. Such changes in the neutron background level were observed during the initial studies of this work.

The following observations in relation to blank measurements (measurement of a non-active waste drum with matrix material) were made based on the campaign of simulated waste items:

1. The practice of performing a blank measurement daily is of upmost importance as this measurement is needed to make a valid "sample minus blank" operation on the measured quantities (R_1 , R_2 , and R_3) to determine the net response of the waste, and to establish a counting regime that allows the standard statistical treatment considering that part of the neutron blank originate from relatively rare neutron bursts (of high multiplicity) produced in the atmosphere by high-energy cosmic particles. The daily blank measurement takes into account the day-by-day variation in the general background level (influenced by pressure, humidity, cosmic ray induced neutron etc.).
2. The daily blank measurement must be representative of the matrix material to be measured same day. This takes into account the background variation due to neutron production and absorption in the matrix material. The operator should take care in the daily planning that the matrix material of the blank measurement corresponds to the waste items to be measured that day. If possible, the blank measurement can be done over night before the sample measurements.
3. Both the daily blank measurements and the sample measurement should be longer than the 1800 s adopted in the present work. This is necessary due to the relatively low count rate in the neutron detectors. This is particularly true for the triples count rate (R_3). The measurement time must be long enough that the observed uncertainty of R_3 is less than 2%. If this is achieved, the uncertainty on R_1 and R_2 will be smaller still.
4. The neutron correlation analysis (analysis of signal singlets, doublets and triplets) is particularly sensitive to electronic noise in the detection system. The reason is that in the neutron correlation analysis, events of repeated electrical noise signals in short time intervals, are indistinguishable from bursts of neutrons. Exactly the bursts of neutrons (from spontaneous fission) are the object of measurement in this analysis method. For this reason, it is of paramount importance that the detection system is noise free, and carefully adjusted to discriminate electrical noise, gamma rays etc. The operator should check that the blank count rates (R_1 , R_2 , and R_3) do not deviate significantly from previous measurements. Electrical motor drives are known to produce electrical noise visible in the blank measurement.
5. In the present campaign we used the programmable TDS32 as mixer unit for the 18 parallel counting chains of the fission neutron counting system on the device. An artificial dead-time of 1 μ s was introduced to suppress electronic noise in the detection system. This is only acceptable because the counting rates during these waste measurements are comparable to the background level. At higher count rates this solution would not be acceptable, as a bias would incur. In the long term, a better solution would be to substitute the detector front-end electronics (pre-amplifier, amplifier and discriminator circuits) with noise-free components.

In relation to the measurements of simulated waste, the following observations were made [20]:

6. The application of the neutron correlation method is a good solution for the passive neutron measurements of waste for matrices of relatively low neutron moderation and absorption properties. The logical method to apply for the analysis of the measured R_1 , R_2 , and R_3 is to solve for the spontaneous fission rate, the average neutron detection efficiency, and the alpha ratio, while assuming no neutron self-multiplication in the waste ($M = 1$). The standard practice, in nuclear safeguards of bulk material, to assume a known detection

efficiency does not work due to the efficiency dependence on the location within the waste matrix.

7. The passive/active neutron station of WCS is designed as a compromise between a dedicated passive neutron detection system (with many ^3He neutron detectors arranged in 4p geometry) and a Differential Die-Away detection system (with a large graphite mantle in front of the fission neutron detectors). Due to this compromise, the neutron detection efficiency (for spontaneous fission neutrons) in centre of the empty cavity of 12.2%. This is rather low for a stable measurement of signal triplets (R_3), particularly taking into account the any matrix material will inevitably reduce the efficiency further. In principle a dedicated instrument such as the JRC Waste Drum Monitor (not available in this campaign) better serves passive neutron measurements due to the much higher neutron detection efficiency (of 30%). The low detection efficiency of the passive/active neutron station can to some extend be compensated by selecting long measurement times. However long measurement times also increases the possibility of suffering from electronic noise, and even the relatively rare cosmic ray induced neutron bursts. It is recommended that the total measurement time be longer than the 1800 s generally applied in this study, and to subdivide the measurement time into 100 s repetitions. The acquisition system must update the average values (of R_1 , R_2 , and R_3) following each repetition, and remove outliers caused by spurious neutron or electronic noise bursts after each repletion.
8. The measurements in the matrices: air, metal, technological non-combustible, technological combustible, and cement showed that the chosen measurement time (typically 1800 s) could with benefit have been chosen longer, preferably 3600 s in general, and 7200 s for the most hydrogenous matrices (cement, and technological combustible). For the three least-hydrogenous matrices, the assay result agreed within 2-3 standard deviations with the known Pu mass of the standards.
9. In the case of the cement matrix a stable measurement of the signal triplets (R_3) above the background variation, is not achieved for the ^{240}Pu masses applied in this study. The low values observed, are due to R_3 being proportional to the efficiency in the third power. In this case, only the measured R_1 , R_2 , quantities can be used to determine the $^{240}\text{Pu}_{\text{eff}}$ mass. As solution is recommended to either solve the equations for R_1 and R_2 directly (see [21], or to calibrate the measured R_2 values against mass of $^{240}\text{Pu}_{\text{eff}}$ using known Pu standards and maintaining the assumption of no self-multiplication ($M=1$) (see Chapter 4). In either case, the discrepancy between assayed and real mass is can exceed 50% mainly due to the variations of the neutron detection efficiency for Pu distributed in the waste matrix.
10. As the passive neutron method determines the mass of $^{240}\text{Pu}_{\text{eff}}$ only, the knowledge of the isotopic abundances of Pu is needed to achieve the mass of total Pu. The isotopic composition can be estimated from the measurement at the TGS/SGS gamma station. Alternatively, an assumption of the ^{240}Pu abundance can be made based on the estimate/knowledge of the Pu burn-up of the material in the waste. This however introduces a further uncertainty in the total Pu mass determination.
The neutron correlation measurement essentially determines the Pu mass through the determination of the spontaneous fission rate of the waste item. This assumes that only Pu isotopes contribute to the spontaneous fission. Other actinides, such as ^{244}Cm , have significantly higher specific spontaneous fission rates than the Pu isotopes. If such isotopes are present in the waste, the Pu mass cannot be determined by passive neutron assay. In such cases only the active neutron assay (DDA technique) can achieve a Pu mass estimate by means of thermal neutron induced fission of the ^{239}Pu and ^{241}Pu isotopes.

In relation to the measurement campaign on drums of real waste [24], the following observations were made:

11. All real waste drums were measured three to four times. These measurements were typically separated by several days. Thus any repetition cycle included a separate blank measurement, as well as the drum movements and positioning in the drum cavity. The 3-4

repetitions gave identical measurement values (R_1 , R_2 and R_3) within the measurement uncertainty, thus demonstrating good reproducibility.

12. Only for two real waste drums of type TC, a net neutron count rate (R_1) significantly above the blank value was observed. These were the drums TC2 and TC4 with the R_1 values $1.980 \pm 0.045 \text{ s}^{-1}$ and $1.176 \pm 0.047 \text{ s}^{-1}$, respectively. For both drums the R_2 and R_3 were zero or negative, indicating the absence of spontaneous fissile material above the MDA level. The two drums however have a net neutron emission rate to be attributed to another kind of source. The likely candidate is (α , n) reactions of an α -emitter in conjunction with light material. A possible candidate could be $^{241}\text{AmO}_2$. The presence of ^{241}Am had already been observed in some drums at the gamma scanning station. The measurement of the singlets rate alone, does not allow for quantification.

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doi:10.2760/748464
ISBN 978-92-76-40051-6