Characterization of legacy waste drums containing UF_6 by Differential Die-away Technique

Stefano Vaccaro
European Commission, Joint Research Centre, Ispra, Italy
Marta Cerini, Ph.D.
Bouygues Construction Expertises Nucleaires c/o European Commission JRC - Ispra
Matteo Mazzuccato, Ph.D.
European Commission, Joint Research Centre, Ispra, Italy
Paolo Peerani
European Commission, Joint Research Centre, Ispra, Italy

Abstract

Radioactive waste drums containing nuclear material pose a significant challenge in the context of JRC-Ispra decommissioning and waste management, both for the alpha emitting radionuclide content and for the safeguards implications. Accurate characterization of these drums is therefore essential for the safe management of nuclear waste.

In this paper, we describe the use of differential die-away technique neutron measurements (DDT) and gamma spectrometry for the characterization of radioactive waste drums containing Uranium Hexafluoride (UF_6). Our results show that the combination of these techniques provides a highly accurate and sensitive means of characterizing these drums, including the mass and the FRAM-evaluated enrichment of Uranium, as well as the consequent alpha activity.

We also discuss the general benefits of using DDT in waste characterization, specifically the ability to non-invasively detect and quantify the fissile content of the drums. Our findings highlight the importance of nuclear material characterization in the management of nuclear waste.

---

1. Introduction

European Commission's Joint Research Center site in Ispra (Italy) has been a nuclear research center since 1959. It still plays a fundamental role for the European Commission science-based policy support, as the biggest site of the European Science Hub, while, during the 1980's, the nuclear research program was progressively reduced and the nuclear research facilities shut down. The nuclear part of the center is currently undergoing a multi-annual Decommissioning and Waste Management Program, to progressively eliminate historical liabilities.
Significant quantities of radioactive wastes were accumulated on site either during the operational time or afterwards, eventually resulting in a plethora of different waste forms: loose items, in-drum or retained waste, waste conditioned in bitumen or cement, waste disposed of in trenches or pits [1]. The radiological features of the various items, need to be accurately assessed to properly classify the material in the categories defined at regulatory level and carry-out the necessary pre-disposal activities. To this purpose, the JRC Nuclear Decommissioning and Waste Management Directorate (JRC.J) has acquired competence and cutting-edge technologies, to respond to the characterization needs at the level required to minimize environmental risks, thus obtaining the necessary public support. In this framework, the Radioactive Waste Management Facility was equipped in the early 2000s with an automated Waste Characterization System (WCS), allowing gamma-ray spectrometric characterization by Segmented and Tomographic

Gamma Scanning, as well as Neutron Active and Passive assay for fissile content determination [2]. The system is at present undergoing a major update and maintenance intervention, to cope with technological aging and to optimize the system itself to current needs.
Over its nuclear history, the JRC Ispra site has also played a fundamental role as training and research and development facility in support of inspectorates for the verification of the Euratom and Non-proliferation Treaties [3][4]. Both in the context of the European Commission (the Euratom safeguards inspectorate, now in DG ENER) and for international organizations, like the International Atomic Energy Agency, the JRC has established many partnerships which brought to important advancements in the knowledge and the technologies specific of Nuclear Material Management and Verification.
This activities result now in the presence, within the legacy waste, of nuclear material from old inspectors' verification samples, gathered along the years until the end of the 1980s, thus scarcely documented. The present paper describes the characterization of two of those samples' collections in small 60 liter drums containing several UF_6 cylinders, carried out by active neutron Differential Die-Away Technique and Gamma-ray spectroscopy, finally resulting in a more accurate update of the NMAC for the concerned Material Balance Area.

2. Materials and methods

2.1. Gamma assay

Open geometry gamma-ray measurements, with a continuous rotation of the drum, were performed to acquire the gamma spectrum of the small drums and samples in order to determine the enrichment of ^{235}U. The enrichment in ^{235}U was determined analyzing the gamma-ray spectra by FRAM (Fixed-Energy Response-Function Analysis with Multi-Efficiency) software [6]. In this case, the ORTEC D-Spec 50 electronics, connected to the Ortec SGD-GEM-5018040-P41038A HPGe gamma detector, was coherent with the FRAM settings.

2.2. Neutron assay

2.2.1. DDT measurements at WCS

The small drums were inserted inside 220l drums using a centring assembly and in this configuration they were measured inside the neutron detector cavity (Figure 1).
The neutron cavity is characterized by 18 banks of ^{3}He detectors: three banks for each side of the cavity. In each bank, the ^{3}He detector tubes are embedded in a block of high density polyethylene which cover the full height of the cavity. Each detector bank is covered in a cadmium liner on all surfaces except towards neighbouring banks. Each detector bank has an electronic junction box connected to each ^{3}He detector. Each junction box includes a preamplifier which sends a TTL signal in a mixer-de-randomizer. The mixer circuit collects all 18 signals coming from each detector bank and feeds a single signal pulse which is fed into a signal analyser directly connected to the acquisition software.

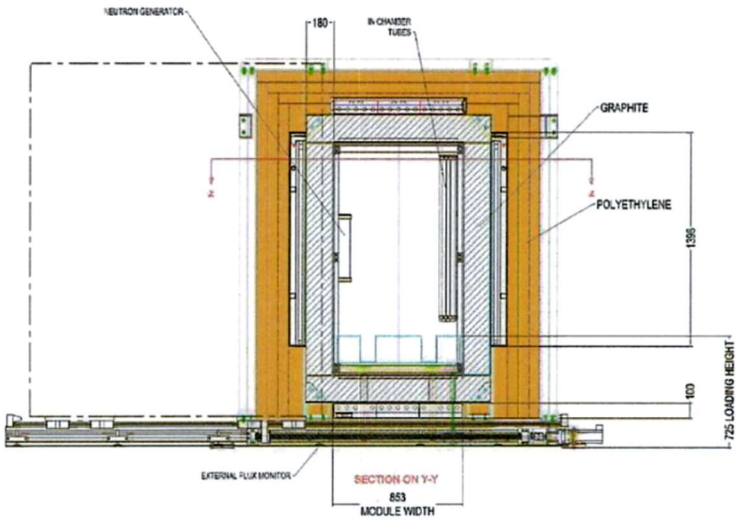

Figure 1 Neutron detector schematic [2]

For neutron active measurements, the calibration was carried out with continuous rotation of the drum, by interrogating the sample using a Zetatron-type neutron pulsed generator system model P-211 produced by Thermo MF Physics, showed in Figure 2, with the following settings:

- Max rate 100 Hz,
- Preset count at 50000 (it means 8 minutes for each measurement),
- Target voltage at 400 V,
- Source voltage at 500 V

Figure 2 Zetatron-type neutron pulsed generator system model P-211

To assess the results dispersion, thus the measurement uncertainty, four repetitions of the small drums measurements were carried out. For each repetition, the normalized counts are evaluated according to (1) [7]:

$$Normalized\ counts = \left(\frac{EG-LG}{SM}\right) \qquad (1)$$

where:

- EG is the early gate and it is obtained from 0.8 ms to 3 ms;
- LG is the late gate and it is obtained from 6 ms to 8.2 ms;

- SM is the source monitor normalization value which takes into account the generator source variability.

The real counts, corrected for matrix effect by means of on barrel monitor, are then reported in equation (2):

$$Real\ counts = Normalized\ counts \cdot OBM_{corr} \qquad (2)$$

where:

- Normalized counts are determined from equation (1).
- OBM_{corr} is the on-barrel monitor correction and it is obtained from 0.35 to 4.8 ms and normalized with the source monitor. It is given by the ratio between the OBM value of air drum and the one of the matrix reference drum.

The three MCS patterns are observed from MCS-32 and remotely controlled from the control room and are obtained from:

1. Fission neutron counters which are directly connected to 18 pre-amplifiers of ^{3}He tubes of the neutron chamber. This spectrum is characterized by:
    a. Early Gate (EG), which is due to thermal source neutrons including fission in fissile material with subsequent detection of the fast prompt fission neutrons.
    b. Late Gate (LG), which is the signal that is un-correlated to the triggering event.
2. Source monitor (SM), which is outside the measurement cavity and it only detects the source neutrons produced from neutron generator tube.
3. On barrel monitor (OBM), installed inside the detector cavity very close to the drum, so that it only detects the direct neutron flux coming from the drum. The OBM takes into account the moderation (from 0.35 ms to 1 ms) and absorption effects (from 1.0 ms to 4.8 ms).

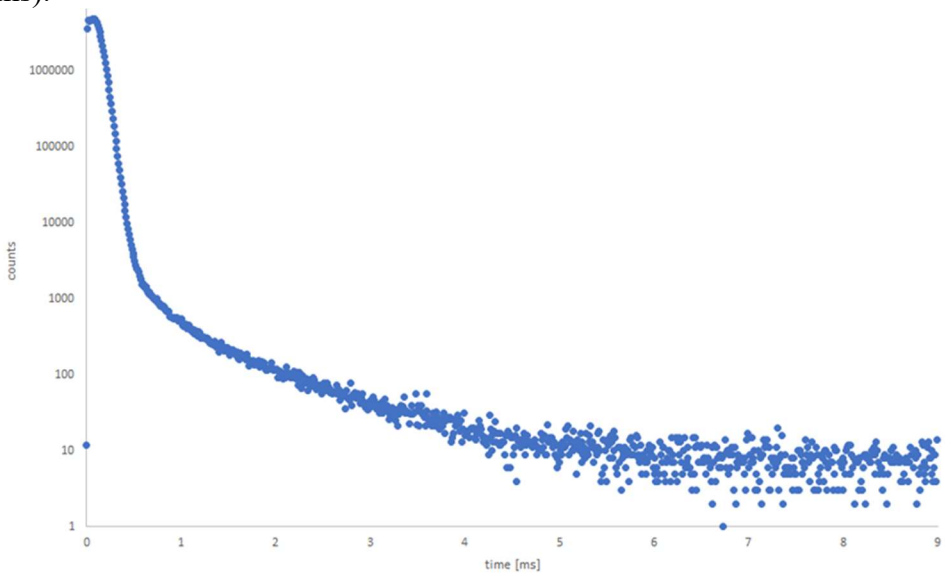

Figure 3 MCS acquisition pattern of the neutron counts from the detector cavity

2.3. Calibration materials

This paragraph lists the materials used to calibrate the DDT system for the Uranium measurements. The standards set consists of two distinct subsets: the CBNM-171 U_3O_8 powder reference materials and a set of laboratory-prepared working standards for ^{235}U masses below 500 mg.

2.3.1. CBNM 171 reference materials

The Nuclear Reference Material 171 (referred to as CBNM-NRM-171 or EC-NRM-171) is a set of low-enriched uranium standards [5] which was jointly prepared and certified by the former Central Bureau of Nuclear Measurements (CBNM) (presently JRC Geel) and the former National Bureau of Standards (NBS) of the US (presently National Institute for Science and Technology (NIST)). A CBNM-NRM-171 set consists of five sealed aluminum cylindrical cans, each containing 200.0 (+/- 0.2) g of U_3O_8 pressed powder, with a different ^{235}U abundance. They are labelled according to their nominal relative amount of ^{235}U.
The isotopic composition and the relative uranium content are reported in *Table 1*. From these data and the Uranium mass the mass of fissile ^{235}U can be calculated and is displayed in the last column.

Table 1 Isotopic composition, relative U content, ^{235}U mass

SAMPLE ID	$^{234}U/U$	$^{235}U/U$	$^{236}U/U$	$^{238}U/U$	U/U_3O_8	^{235}U
	at%	at%	at%	at%	wt%	g
CBNM-031	0.0020 (2)	0.3206 (2)	0.0147 (3)	99.6627 (4)	84.56 (15)	0.542 (3)
CBNM-071	0.0053 (2)	0.7209 (5)	<0.00002	99.2738 (4)	84.53 (15)	1.219 (3)
CBNM-194	0.0174 (2)	1.9664 (14)	0.0003 (1)	98.0159 (18)	84.56 (15)	3.326 (3)
CBNM-295	0.0284 (4)	2.9857 (21)	0.0033 (2)	96.9826 (29)	84.52 (15)	5.047 (3)
CBNM-446	0.0365 (3)	4.5168 (32)	0.0069 (2)	95.4398 (32)	84.66 (15)	7.648 (3)

Figure 4 shows the resulting calibration curve for high ^{235}U masses. It appears a clear self-attenuation effect above 5 grams, with consequent under-linearity of the curve.

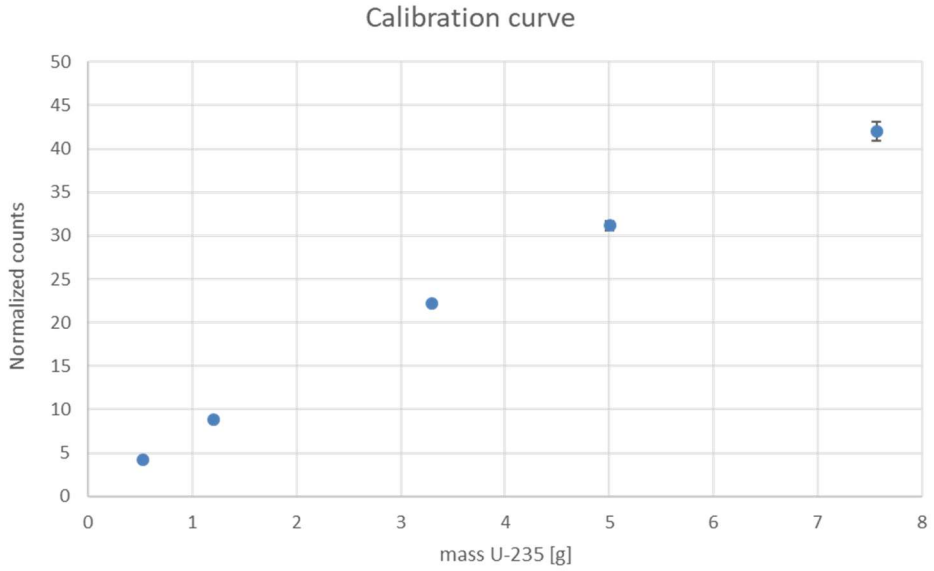

Figure 4 Calibration curve for ^{235}U masses above 0.5 grams

2.3.2. Working standards

The ^{235}U mass from CBNM reference materials (the lowest is 542 mg) does not allow to have a calibration curve properly working in the ^{235}U mass range from 0 to 0.50 g, where the vast majority of U-bearing waste drums stands.

Therefore, the DDT has been calibrated with additional natural Uranium working standards, produced on purpose in laboratory, with mass ranging from 0 mg to 500 mg in order to have a calibration curve that allows to determinate ^{235}U mass from tens of milligrams to grams combining with the calibration curve obtained from CBNM reference materials. From the complete calibration curve, we obtain:

- intercept of the calibration curve because the blank measurement is not zero,
- the minimum detectable amount of ^{235}U,
- a small quantity of ^{235}U in nuclear waste drums with satisfactory accuracy.

Table 2 Reports the mass of ^{235}U in the working standards set used and Figure 5 represent the resulting calibration curve. For those low masses the curve appears strongly linear, with no detectable self-attenuation.

Table 2 ^{235}U Mass in the working standards

SAMPLE ID	^{235}U MASS
WS 1	0.026 g
WS 2	0.054 g
WS 3	0.217 g
WS 4	0.527 g
WS 5	1.207 g
WS 6	3.298 g

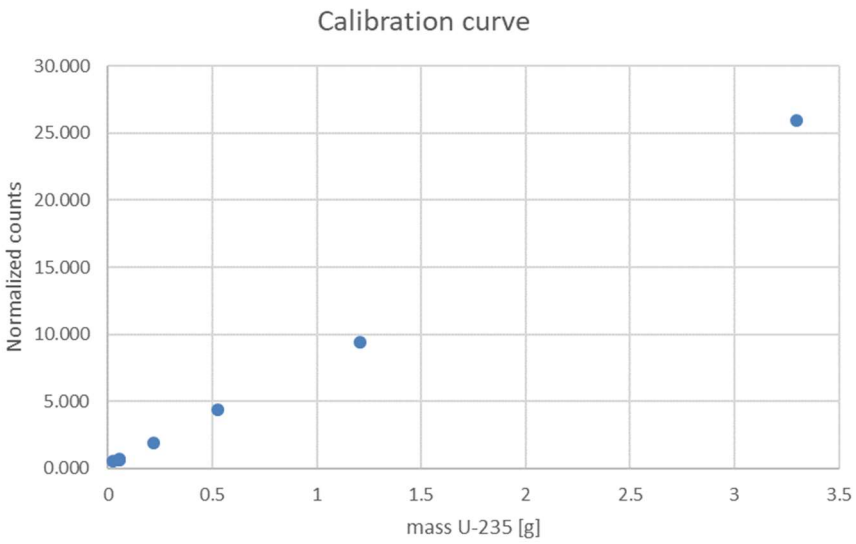

Figure 5 Calibration curve for low ^{235}U masses

3. Results

The two small drums have been were obtained during a sorting and repackaging campaign in the Radioactive Waste Management Facility of JRC Ispra. The characterization measurements consisted in:

- Neutron Differential Die-Away Technique – DDT;
- Gamma spectroscopy for isotopic composition determination;

The 60 litres small drums have been inserted in standard 220 litres drums by means of a centring assembly, for mechanical compatibility, in order to perform the investigations in automated facilities.

The small drums contain all the aforementioned UF_6 containing metal bottles from legacy nuclear inspection samples. The UF_6 bottles were wrapped in two plastic bags in order to avoid any α-contamination in the event of future inspections.
The small 60 litres drums after the repackaging treatment (separating UF_6 bottles items from other waste) consist of:

- Drum 1: small drum containing 38 items;
- Drum 2: small drum containing 31 items;

The two objects underwent DDT measurement at the WCS, to determine the amount of fissile material contained, namely ^{235}U, and accurately assess the fissile isotope mass, the key parameter according to Euratom regulation 302/2005. Subsequently, a gamma-ray spectrometry measurement was performed, in order to determine the Uranium average enrichment, hence the heavy metal total mass. The spectra are analysed with FRAM.

3.1.Drum 1

The measurement was performed as described in §2.3.1. The real counts of Drum 1 were obtained according to equation (2) by considering the drum matrix as a mixture of air, metal and technological combustible (plastic).
To assess the results dispersion, thus the measurement uncertainty, four repetitions of the small drum measurement were carried out. The resulting normalized and matrix-corrected counts are *25.7907 ± 0.8776*.
According to the calibration in paragraph 2.3.2 this corresponds to ^{235}U mass of *2.99 ± 0.08 g*, that gives an α–activity of this radionuclide of *239.52 ± 6.62 kB*q.
The measured average enrichment of ^{235}U in Drum 1 is *2.58 ± 0.83%,* therefore, the total mass of U in the small drum is *116.04 ± 37.47 g*, this quantity is derived combining the mass and the enrichment of U-235 in the drum.

3.2.Drum 2

Also for this drum, the same characterization procedure as for Drum 1 was applied.
The normalized and matrix-corrected counts of drum 2 result in *14.5349 ± 0.8587*, corresponding a ^{235}U mass of *1.63 ± 0.07 g*.
From the mass of ^{235}U, the activity of this radionuclide is *130.13 ± 5.56 kB*q.
The measured average enrichment of ^{235}U in the drum is *1.61 ± 0.61%,* therefore, the total mass of U in the small drum is *101.01 ± 38.52 g*.

4. Conclusions

Two small drums, reportedly containing UF_6 samples from safeguards inspection, were measured by DDT and gamma spectroscopy by JRC Ispra Waste Characterization System. The DDT detected the presence of fissile material, ^{235}U. The gamma spectroscopy allowed to determine the ^{235}U enrichment.
According to EU directive 302/2005, the key parameter for NMAC is the mass of fissile material, in this case ^{235}U. This work describes how a DDT assay ensures that the mass values can be obtained very accurately, by taking into account the absorption and moderation effects cause by the matrix with a suitable detector configuration, down to the order of magnitude of tens of milligrams.

References

[1] Brunetti, Gianfranco, Accorsi, Roberto, Aulamo, Heikki, Doublecourt, Jean-Jacques, Guisset, Jean-Philippe, & Zanovello, Flavio (2013). Historical Radioactive Waste Management at the JRC-Ispra Site - 13059. 5 International Conference and Exhibition on Decommissioning Challenges - Industrial Reality and Prospects, France

[2] J. A. Mason, W. Hage, R. Price, A. C. Tolchard, and A. C. N. Towner, "An Automated Non-Destructive Assay System for the Measurement and Characterization of Radioactive Waste," Proc. Ninth International Conference on Radioactive Waste Management and Environmental Remediation (ICEM'03), Oxford, England, October, 2003.

[3] Consolidated version of the Treaty establishing the European Atomic Energy Community, 2012/C 327/01, https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A12012A%2FTXT

[4] Treaty on the Non-Proliferation of Nuclear Weapons (NPT) https://www.iaea.org/publications/documents/treaties/npt

[5] Paradela, G. Alaerts, J. Heyse, S. Kopecky, A. Moens, P. Schillebeeckx, R. Wynants, H. Harada, F. Kitatani, F. Ma, H. Tsuchiya, Reference samples for NRA applications, EUR29977 EN, Publications Office of the European Union, Luxembourg, 2019, ISBN 978-92-76-13090-1, doi:10.2760/448850, JRC118422

[6] Vo, Duc T., and Sampson, Thomas E. FRAM, Version 6.1 User Manual. United States: N. p., 2020. Web. DOI:10.2172/1599022

[7] ASTM C1493 − 19 Standard Test Method for Non-Destructive Assay of Nuclear Material in Waste by Passive and Active Neutron Counting Using a Differential Die-Away System, ASTM, 2019. DOI: 10.1520/C1493-19

[8] Commission Regulation (Euratom) No 302/2005 of 8 February 2005 on the application of Euratom safeguards - Council/Commission statement (OJ L 54 28.02.2005, p. 1, ELI: http://data.europa.eu/eli/reg/2005/302/oj)