Quantitative Radiological Characterization of Waste: Integration of Gamma Spectrometry and Passive/Active Neutron Assay

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The paper "Quantitative Radiological Characterization of Waste: Integration of Gamma Spectrometry and Passive/Active Neutron Assay" by Dr. Gianluca Simone, Dr. Egidio Mauro, Dr. Filippo Gagliardi and Dr. Edoardo Gorello has been awarded as Best Paper of the 47th Annual Meeting on Nuclear Technology (AMNT 2016), Hamburg, 10 to 12 May 2016.

Introduction In order to determine the full physical inventory of nuclear material in drums (both for safeguard purposes and for classification/transportation/storage) several assay methods are required, in particular if the drums matrix is not well known. In fact, all the possible methods have some limitations, which must be understood and corrected. For example, the widely-used gamma spectrometry (potentially able to detect all the relevant uranium and plutonium radioisotopes) has the issue of the photons absorption due to the matrix and to the nuclear materials.

The matrix absorption can be evaluated through Monte Carlo calculations (if the details of the matrix can be inferred) or through a density measure (using a transmission source). Usually, a characterization system evaluates automatically the measurement efficiency (and so the matrix absorption). On the contrary, the self- absorption due to the nuclear material can be estimated only knowing its spatial distribution and total amount – which is one of the goals of the gamma spectrometry – and has to be corrected a *posteriori*. This effect is more and more severe as the SNM mass increases.

Some information about the mass of the nuclear material can be obtained from historical data (if any) or using other methods, like a neutron counting assay. In this way, the results from the gamma spectrometry can be compared to those from an independent method and, if an important discrepancy emerges, can be corrected for the self-absorption effect.

The selected techniques

To carry out a full and trustable ND radiological characterization of drums, *Nucleco* has implemented several *CANBERRA* assay systems:

- The NWAS (Nucleco Waste Assay System), which performs a segmented high resolution, gamma scanning – SGS – on 220 ℓ drums;
- Several ISOCS-like systems (i.e. far-field high-resolution gamma spectrometers), adequate for almost every kind of material and geometry. The system, called MGAS – Mobile Gamma Assay System – is dedicated exclusively to drums of different volumes;
- An active/passive neutron assay system (PANWAS – Passive Active Neutron Waste Assay System) *[1]*.

These systems create an integrated gamma/neutron waste characterisation facility. These instruments feed a common database so that, when items are measured on more than one station, a consolidated record is available for analysis. The choice of the mentioned techniques (gamma and passive/active neutron assay) is based on the specific problems related to the evaluation of uranium and plutonium radioisotopes.

Depending on the geometrical efficiency and the statistics, potentially all the main radioisotopes of uranium (U-235, U-238) and plutonium (Pu-238, Pu-239, Pu-240, Pu-241) are detectable through gamma spectrometry. However, this assay can be affected by a severe self-absorption phenomenon (in particular in the energy range up to few hundred keV), which may cause wrong or inconsistent results. The self-absorption becomes more and more important as the mass of the nuclear material increases, affecting also the assay at the energy of MeV when the mass of kilograms are considered.

About the passive neutron assay, even mass number plutonium radioisotopes (Pu- 238, Pu-240) have a high spontaneous fission probability, making the neutron emission rate detectable in passive counting system. On the contrary, U-238 is generally considered not to be detectable by passive techniques because of the low spontaneous fission neutron emission rate. However, a quantity of U-238 greater than approximately 1 kg emits a measurable spontaneous fission neutron signal. For this reason quantities of U-238 lower than about 1 to 10 kg can be detected only by gamma assay (with good accuracy) while, for higher quantities, passive neutron assay can be used as an integration of the gamma assay (affected by self-absorption).

About the active neutron assay, odd mass number Pu radioisotopes (Pu-239, Pu-241) and U-235 have a cross section for induced thermal fission high enough to be detected through this technique, usually with a sensitivity starting from few mg.

These measurement systems allow to compare and integrate the results from different and independent techniques, and let phenomena as self-absorption to be detected and corrected. Methods or algorithms for the automatic correction of the gamma self-absorption are not generally implemented in the analysis software used for these kind of measurements because without expert review they can be misleading. In spite of the fact that the self-absorption and the matrix absorption are physically the same kind of process, the methods commonly used to correct for the presence of the matrix are not sufficient for self-absorption.

The SGS system divides the drum in 8 segments in order to evaluate the axial source and density distributions with a reasonable good approximation. The system evaluates the efficiency using a set of calibrations based multi-curves – SUM mode – (efficiency for the entire drum vs. energy as a function of matrix density), which can be very precise in case of homogeneous matrix, or using an external Eu-152 source to compute the transmission factor for each segment – NID mode –. Nevertheless, when the SNM is concentrated in a small volume its effect (high density, high-Z, K-edge) cannot be detected using neither multi-curve nor external source, implying an underestimation of the emitted photons. Unfortunately, in waste assay an accurate knowledge of the geometry and of the chemical/physical properties of the source is seldom available.

The integration of the results obtained by the three assay methods (neutron counting, gamma spectrometry in segmented and open-geometry modes) is a very important

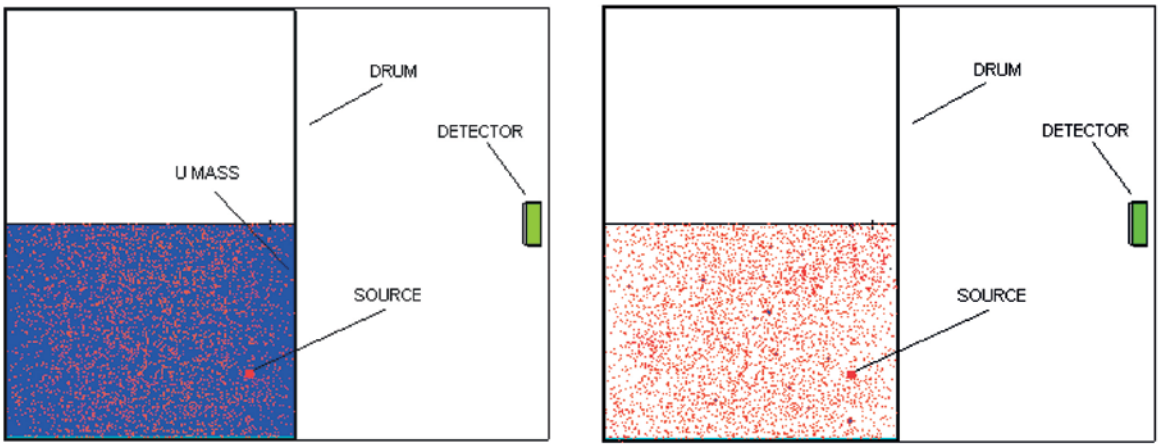

Fig. 1.
Schematic drawing of the two MCNP simulations.

issue since a disagreement between the outcome of the techniques can happen for the reasons described above. In order to allow the integration of the gamma and neutron results for high SNM mass, the simulation code MCNP™ (Monte Carlo N- Particle Transport Code) has been used to calculate the gamma self-absorption factor, within limits to which the geometry may be inferred. The reliability of the simulation results depends strictly on the accuracy of the input data. Unfortunately, in ND assay it is very difficult to know with high accuracy the relevant information (geometry, distribution, chemical and physical form, history) for both radioactive source and matrix.

Description of the simulation

MCNP-4c code *[2]* was used to simulate the gamma measurements for the calculation of the self-absorption correction factor to be applied to the gamma results.

The following procedure has been developed, tested and applied on drums containing a high uranium mass: the drums (220 ℓ, in general containing depleted U) have been chosen since the gamma and neutron assays, even though discordant, confirmed the presence of a high U mass.

The geometry used for the simulation is described schematically in **Figure 1** – *"Schematic drawing of the two MCNP simulations"*. The axial distribution of the source (dots) comes from the results of the segmented gamma assay, while the radial distribution is assumed homogeneous. For all drums containing a high U mass, two different simulations have been performed: in the first one the volume in which the source is distributed (the source volume) is filled with uranium mass (blue dots); in the second one the same source volume is assumed to be filled with air. The matrix is not taken into account, since the experimental spectrum to which the simulation will be compared is already corrected for the gross matrix effects.

Given N and N' respectively the number of gamma simulated in the first and in the second step:

$$N = N_{tot} \cdot \varepsilon \cdot CF$$

$$N' = N_{tot} \cdot \varepsilon$$

$$CF = \frac{N}{N'}$$

$$CF = CF_{Z,K} \cdot CF_{\rho}$$

where N_{tot} is the total source gamma emission and is the measured geometrical efficiency. The parameter CF is the correction factor due to the uranium itself, which depends both on the mass density distribution (corrected by CF_{ρ}) and on the high-Z effect and the K-edge absorption (corrected by $CF_{Z,K}$). **Figure 2** *"Comparision of inverse absorption correction factor for several uranium mass and drum filling"* shows the absorption correction factor for U-238 (U-235) for several U-238 (U-235) masses and drum filling heights.

Using Monte Carlo simulation, both $CF_{Z,K}$ and CF can be evaluated. However, in order to use one or the other for the integration procedure, some attention must be taken: using the first one $CF_{Z,K}$ means that the correction for the mass density distribution is entrusted to the multi-curve method, while using CF implies that correction is done both using simulation and multi-curve. Thanks to the segmented scanning, the uranium mass distribution is known with good precision so the best approach is to use CF and to recalculate the multi-curve efficiency excluding the uranium mass (taken from the PANWAS) in the calculation of the drum apparent density.

Integration procedure and results

The results of the simulation procedure described above allow a comparison and self-validation of the results coming from the gamma and

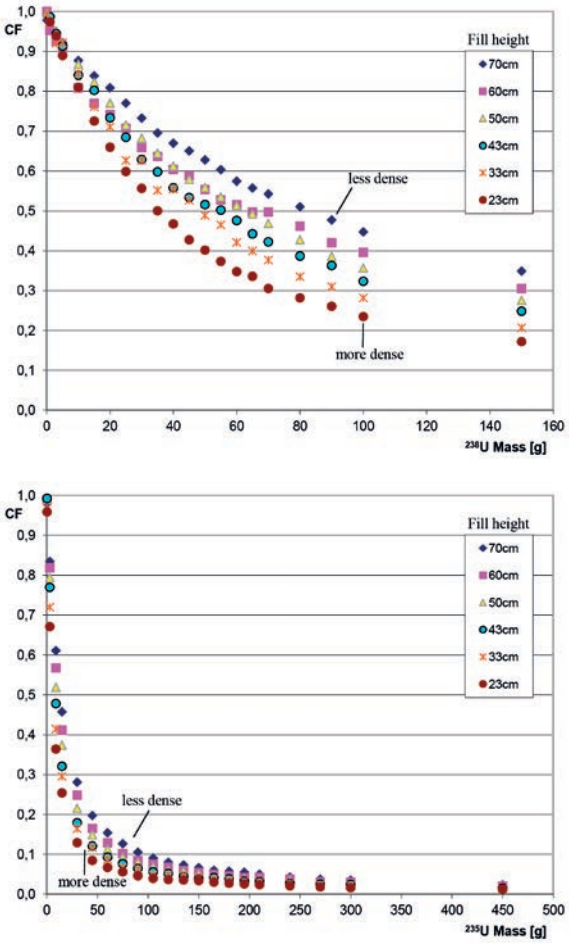

Fig. 2.
Comparision of inverse absorption correction factor for several uranium mass and drum filling.

the neutron assays. **Table 1** *"Results of the quantitative gamma-neutron integration procedure"* summarizes the results of the integrated quantitative neutron-gamma procedure for 32 more relevant drums.

By taking the uranium mass from the neutron assay (columns 5, 6), the source distribution from the gamma-scanning assay (column 3), the gamma self-absorption correction factor C_{abs} for U-238 (U-235) is calculated through MCNP™ simulation. Applying this factor to the U-238 (U-235) mass measured from SGS – SUM mode – (columns 7, 8) and recalculating the multi-curve efficiency to the "uranium-less" mass density, the corrected U-238 (U-235) mass is computed (columns 10, 11). This mass, afterwards, has been compared to the one measured by the neutron assay (columns 13, 14).

As it is clear from **Tab. 1**, the agreement between neutron and gamma seems better as more as the uranium is the main component of the drum matrix. In fact, if U is the main matrix component, the average-density effect is well described both in SGS and in MCNP™ simulation.

An agreement better than 25 % is achieved for U-238 in most of the drums, especially for those containing depleted uranium and named "U_{dep} Spezzano". In these cases, the simulation is particularly accurate since the source is uniformly distributed, as confirmed by the historical information. Moreover, for these drums uranium is the main contribution to the total mass. However, U-238 suffers a relatively low self-absorption effect and the general good agreement is a further confirmation about the accuracy of the method. For the remaining drums, the higher deviation can be attributed to a non-homogeneous uranium distribution.

A similar behaviour is observed for U-235. Quite relevant is the fact that for gross mass higher than 150 kg the corrected gamma result is heavily higher than the neutron result. This can be due to several factors, like the U-235 mass distribution or the application of the NWAS and PANWAS calibrations near to the limits of the systems. It's also important to keep in mind that a possible under-corrected self- shielding *[4]* of the active neutron signal could justify these discrepancies. Such cases will be further studied.

It must be noted that in those cases where is not possible to reconcile the gamma and neutron results, the quoted result is always the one coming from the neutron assay for the above

N°	Drum information			Neutron assay		Non corrected gamma assay		Corrected gamma assay		Discrepancy	
	Gross weight [kg]	Source height [cm]	Historical information	U-238 mass [kg]	U-235 mass [g]	U-238 mass [kg]	U-235 mass [g]	U-238 mass [kg]	U-235 mass [g]	U-238 mass	U-235 mass
1	72	20	Sources Unat	0.8	2.4	0.5	1.8	0.6	2.8	-25 %	21 %
2	128	30	Sources Unat	3.4	16.8	2.5	7.7	2.9	19.4	-15 %	16 %
3	78	41	Udep spezzano	53.9	196.6	30.3	14.5	47.8	179.8	-11 %	-9 %
4	78	41	Udep spezzano	53.4	180.9	32.6	15.6	51.4	193.3	-4 %	7 %
5	78	41	Udep spezzano	51.8	180.5	33.8	17.0	53.3	210.4	3 %	17 %
6	78	41	Udep spezzano	53.6	179.7	32.6	16.2	51.4	200.4	-4 %	12 %
7	78	46	Udep spezzano	53.6	178.8	34.6	17.1	51.5	192.8	-4 %	8 %
8	196	46	Udep caesar	30.2	70.2	23.0	38.8	29.9	280.1	-1 %	299 %
9	106	30	Udep caesar	10.7	20.9	5.7	3.2	7.0	20.3	-35 %	-3 %
10	78	41	Udep spezzano	53.9	175.6	33.6	16.5	53.0	204.2	-2 %	16 %
11	78	45	Udep spezzano	51.4	151.3	33.5	16.4	49.7	188.1	-3 %	24 %
12	78	41	Udep spezzano	54.0	167.5	30.8	13.7	49.8	176.9	-8 %	6 %
13	78	48	Udep spezzano	51.8	171.1	33.4	16.1	45.7	161.3	-12 %	-6 %
14	74	35	Udep spezzano	48.6	173.2	28.2	13.0	47.5	177.3	-2 %	2 %
15	78	41	Udep spezzano	53.0	166.4	31.5	14.3	49.4	175.8	-7 %	6 %
16	164	41	Udep caesar	25.7	59.6	16.1	10.5	21.1	92.4	-18 %	55 %
17	80	44	Udep spezzano	52.4	174.0	33.8	17.1	51.0	199.1	-3 %	14 %
18	174	52	Udep spezzano	29.4	63.3	18.7	10.3	23.9	83.5	-19 %	32 %
19	180	44	Udep caesar	28.8	71.4	18.1	13.3	23.8	118.5	-18 %	66 %
20	190	52	Udep spezzano	32.9	68.5	21.0	16.1	26.5	135.7	-20 %	98 %
21	78	41	Udep spezzano	53.5	168.3	31.1	14.1	48.9	173.7	-9 %	3 %
22	172	50	Udep caesar	30.4	66.7	20.8	33.5	26.1	275.8	-14 %	313 %
23	84	30	Udep daytona	6.6	2.0	2.4	1.3	2.7	5.6	-59 %	179 %
24	74	30	Sources Unat	7.1	27.1	3.5	7.2	4.0	32.4	-44 %	20 %
25	68	62	Sources Unat	1.8	5.5	1.3	1.9	1.3	2.6	-24 %	-53 %
26	66	41	Sources Udep	38.9	34.9	12.3	2.0	17.8	22.5	-54 %	-36 %
27	106	30	Sources Udep, Unat	51.5	51.2	5.5	0.6	10.5	9.8	-80 %	-81 %
28	40	35	Sources Unat	1.1	5.9	1.1	1.3	1.2	1.8	8 %	-69 %
29	62	30	Sources Udep, Unat	24.3	28.7	8.8	1.7	15.1	24.7	-38 %	-14 %
30	100	25	Sources Udep	57.9	79.5	17.4	5.4	39.1	105.1	-32 %	32 %
31	166	30	Sources Unat	61.9	85.2	17.2	34.2	31.9	208.3	-49 %	144 %
32	84	30	Sources Unat	3.6	10.4	2.7	4.0	3.0	11.1	-17 %	6 %

Tab. 1.
Results of the quantitative gamma-neutron integration procedure.

mentioned reasons. However, also in these cases, the simulation gives us a useful indication on the reliability of the neutron and gamma measurements, when further and more precise information on the source are missing

Extension of the procedure: ISOCS-only

When detailed information on the sample to characterize are available, the presented procedure can be reduced by using only the gamma assay in open geometry. In *Nucleco*, this kind of assay is entrusted to the ISOCS system by *CANBERRA*.

It has not an automatic loading system of drums and has no predefined geometrical efficiency, so for each analysis it costs more time w.r.t. NWAS system. For each measure, the efficiency is evaluated via Monte Carlo simulation and the analyst can invoke the self-absorption correction including the distribution of the SNM in the sample model.

An example of this application is shown in **Figure 3** *"Discrepancy between the Pu- 239 average activity and the line activity calculated for the best Pu-239 emission lines in a spectrum containing hundreds grams of plutonium and uranium"*, which reports the discrepancy of several emission lines of Pu-239 for a cylindrical sample containing hundreds grams of uranium and plutonium oxide. The activities of best energy lines (i.e. the ones with more statistics and less interferences) agree with the mean value with only a 10 % discrepancy and no serious outlier. Moreover, the discrepancy between the ISOCS results and the other data available for the sample (historical data, destructive analysis) is below 3 % for both plutonium and uranium, meaning that the measure efficiency is consistently described.

Outlook and conclusion

In this work we have described the integrated quantitative neutron-gamma procedure developed at Nucleco for the characterization of drummed waste containing a high uranium mass. The need of this integrated procedure for these kind of drums is due to the self-absorption phenomenon, which strongly affects the gamma measurements and cannot be corrected in a reliable way with the methods commonly used for the study of matrix-related effects.

The procedure is based on the simulation code MCNP. It allows the gamma self- absorption factor to be calculated, within limits to which the geometry of the source may be inferred. The gamma measurements are corrected for this factor and subsequently compared to the results of the neutron assay (passive for U-238 and active for U-235). Applications have given satisfactory results for both U-235 and U- 238, especially in those cases where the source distribution was homogeneous. The cases where a disagreement between the two techniques is observed are likely due to a lack of accurate information on the source geometry and to uncertainties in the measurements. In these cases the simulation results are still useful since they supply a warning of a possible measurement fault.

In case the neutron and gamma are in disagreement, if there are some possible reasons to doubt of the neutron assay results (because, for U-238, the expected value is below the sensitivity of the detector, or because there are other interfering process involved, like the neutron multiplication), then a correction for gamma self-absorption is absolutely needed. Depending on the case, dedicated MCNP simulations can be evaluated instead of a set of modelling equation for the correction vs. the uranium mass.

The integration procedure can be successfully applied to other SNM, such as plutonium. In order to improve the accuracy and the reliability of the simulation, *Nucleco* has equipped its laboratory with a digital X-ray radiography system, for a precise scanning of the matrix, and an NIT (Neutron Imaging Technique) analysis algorithm for the PANWAS system, which provides a low-resolution localization of neutron point sources (if present). Moreover, *Nucleco* is further developing and refining other techniques in the integration of the ones already used. Whenever possible, detailed information about the matrix/source can be used to deeply evaluate the efficiency and the self-absorption using MCNP also for far-field gamma spectrometry (e.g. ISOCS). In all the other cases, a great improvement is expected from a new Transportable Tomographic Gamma Scanner (TTGS), actually in commissioning phase, which will allow a 3-dimensional knowledge of both matrix and source distribution in the drum, ensuring a punctual and very detailed correction of the absorption and self-absorption phenomena.

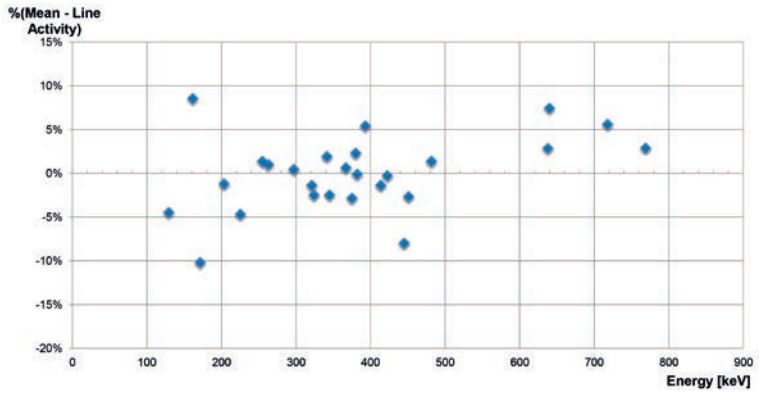

Fig. 3.
Discrepancy between the Pu-239 average activity and the line activity calculated for the best Pu-239 emission lines in a spectrum containing hundreds grams of plutonium and uranium.

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