Calibration verification by Monte-Carlo simulations of a total gamma counting tunnel for clearance purposes

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Abstract

The Joint Research Centre of Ispra is one of the research centres belonging to the European Commission, and was created in the late '50s, in order to steer European research on nuclear industry. It currently hosts numerous nuclear facilities, some of which are maintained in operation, while others were shutdown in the past years or are currently being decommissioned.

The license for the waste station includes clearance for historical and post operations solid materials. As a license requirement for clearance, measurement and characterization of the material must be performed to verify that the radioactive concentration is below the established threshold levels given by the Italian Nuclear Safety Authority.

The final survey on the potentially clearable material homogeneous lots is performed through a commercial total gamma counting tunnel composed by eight plastic scintillators. Calibration is obtained by a standard container through a set of calibration measurements. A verification, via Monte Carlo simulations, of the measurement method of the gamma counting tunnel has been performed in order to have complete reliability on the system used for verifying the radioactive activity in the potentially clearable materials.

Keywords: Material clearance, Monte Carlo, method verification, plastic scintillators.

1. Introduction

The JRC-Ispra manages the Radioactive Waste Management Station (SGRR *Stazione Gestione Rifiuti Radioattivi*), a centralised storage for all historical and post operations wastes generated from the past nuclear activities done on site. The authorisation to operate for SGRR facility is referenced within [1] [2] [3] and later updates and includes clearance of solid materials therein transferred from the six shutdown nuclear facilities. In general, very low level radioactive materials aimed to be recycled, reused or disposed can be released from regulatory control under the condition that the radionuclide concentrations are below the specific clearance levels provided by the Italian Nuclear Safety Authority. Therefore, it is necessary to sort

the material into homogeneous groups, assign to each one of them a well known nuclide vector W[1] and verify that their activity concentrations are below the mentioned clearance levels [4]. So, it has been decided to buy a commercial measuring chain, namely Total Gamma Measurement Chain (TGMC), to perform such verification by total gamma counting on a group of containers statistically representative of the homogeneous group defined by W.

The verification, using Monte Carlo simulations, of the measurement method used by the TGMC has led to the introduction of a safety factor in the global transfer function, in order to take into account possible slight in-homogeneities in the measured containers.

2. TGMC system layout

The TGMC system [5] consists of eight plastic scintillators each coupled with a photomultiplier and disposed to form a tunnel; every scintillator is shielded by lead plates. The passing through container to be assessed is shifted by three successive steps into the tunnel so that only a portion is measured at each step. The activity A_W of the measured container is calculated as the mean of the activities A^d_W measured by each one of the d detectors. The standard procedure for the calibration of the system is performed by a so called standard calibration container filled with steel plates disposed at different levels. Variations on the number of plates at each level allow obtaining different densities for the calibration container. A ^{60}Co planar source placed at different levels represents, by superposition of the effects, a roughly homogeneous distribution of activity in the container for each density. Moreover, a roughly homogeneous distribution of activity for seven mono-energetic nuclides is obtained with point sources symmetrically positioned inside the container.

The TGMC system and the standard calibration container ready to be measured are shown in Figure 1.

Figure 1 - TGMC system and the standard calibration container

[1] The ratios $w_w = \frac{a_w}{\sum_w a_w}$ (between the specific activity a_w of each w radionuclide and the total specific activity of the material) form the nuclide vector W for which is valid the relation: $\sum_w w_w = 1$.

3. TGMC system measurement method

The activities A^d_W determined by the d detector of the TGMC, using the approach given by the supplier, are expressed by [6] [7] [8]:

$$A_W^d = \left(\dot{R}_\Delta^d + z_{sta}^d\right)\frac{f_T^d}{r_W} \quad (1)$$

where $\dot{R}_\Delta^d$ is the net response of the d detector while $\frac{f_T^d}{r_W}$ is the global transfer function for the energies of the radionuclides defined by the nuclide vector W which univocally characterises the material in the container [9] [10]. The correction factors for the photon attenuation in the source f^d_T and r_W are function of the container geometry and material density.

The factor f^d_T is determined using the calibration standard container filled with steel plates disposed on multiple levels. It is calculated by superposition of effects, using a planar ^{60}Co source positioned at such different levels.

The correction factor is then defined by the supplier for eleven densities as:

$$f_T^d = \frac{1}{T_{1.17}\beta_{Co60,1.17} + T_{1.33}\beta_{Co60,1.33}} \quad (2)$$

where T is the transfer function and β the ^{60}Co branching ratios (at 1.17 and 1.33 MeV).

The f^d_T factors, which then apply to all types of material, are obtained only with the mass attenuation coefficient of the iron. This choice is based on the fact that, to the energies of interest where the Compton effect is dominant, there is a negligible dependence between the mass attenuation coefficients of the different materials of interest.

Once the f^d_T correction factor is determined, in order to determine also the r_W correction factor linked to the response at different energies, one (out of seven) point source is placed in sequence in five different positions within the standard calibration container (at the centre of the container and at the centre of each of the four equal sub-volumes obtained by dividing it with two perpendicular planes) and finally the superposition of effects is applied.

The first step to determine the energy response correction factors r_w is to define the following function:

$$\eta(E_i) = \frac{T(E_i)}{T_{1.17}\beta_{Co60,1.17} + T_{1.33}\beta_{Co60,1.33}} \quad (3)$$

determined through the interpolation of η_i values determined for k mono-energetic sources by:

$$\eta_i\beta_{i,k,mon} = \frac{\dot{R}_{k,mon}A_{Co60}}{\dot{R}_{Co60}A_{k,mon}} \quad (4)$$

where activities are known and detector responses measured. Only two configurations of the standard calibration container have been used by the supplier for this calculation: 0.5 g/cm^3 and 2.1 g/cm^3 densities.

The energy response correction factors r_w (or Co-equivalent) for the radionuclide w is then calculated as:

$$r_w = \sum_i \eta(E_i)\beta_{w,i} \quad (5)$$

The correction factor for the whole nuclide vector W of the measured material is then:

$$r_W = \sum_w w_w r_w \quad (6)^2$$

r_w is equal to zero for all the non-gamma emitters and for gamma emitters with emission energy below 300 keV (not detected by the TGMC because of the energy threshold at the optimal photomultiplier high voltage).

As explained before, the energy response correction factors r_w which then apply to all types of material, are obtained only with the mass attenuation coefficient of the iron.

f^d_T and r_W depend on the container geometry and material density. f^d_T is determined for the single d detector while r_W as an average. They are then selected by the TGMC on the basis of the density of the measured container.

The counting ratio z^d_{sta} is included by the supplier and it increases the activity value in order to have, below this value, a true value with a 95% probability.

4. In-homogeneity issue

The system supplier included in (1) the counting ratio z^d_{sta}, but he didn't forecast a safety factor that would take into account the intrinsic non homogeneity of the standard calibration container and the container to be measured. Indeed, the standard calibration container (prepared with steel plates, planar and point sources with non-homogeneous distribution) is only an approximation of an effective homogeneous container. The measured containers then, even if prepared in order to be as much homogeneous as possible, have to be considered only an approximation as well.

So JRC Ispra has investigated the possibility of introducing a safety factor in order to increase the value of the activity determined and to be reasonably confident that such measured value is above the true value. JRC Ispra has used a Monte Carlo model to simulate the TGMC behaviour in two different configurations: (i) with the actual standard calibration container and (ii) with an ideal homogeneous calibration container. In this way it is possible to assess the deviation introduced by the in-homogeneity (assumed as typical) of the standard calibration container and extend it to all the measurements performed by the use of a safety factor.

5. TGMC system measurement method verification by Monte Carlo methodology

A Monte Carlo model [11] [12] of the source-detector geometry has been created and validated (see Figure 2). Once the model has been validated, the following simulations have been executed and compared between them:

- Global geometry with the standard calibration container inside the measurement tunnel;

[2] Note that the transfer functions $T_{1.13}$ and $T_{1.33}$, which depend on the geometry layout, have different values in the two configuration used for the calculation of f^d_T and r_W.

- Global geometry with an ideal calibration container (same density and material of the calibration one, but matrix and source homogeneously distributed).

It has been assumed as typical deviation the one introduced by the actual calibration container (through its in-homogeneity) compared to the ideal one (perfectly homogeneous).

The simulations were performed only at two densities, high and low, respectively 0.5 g/cm^3 and 2.1 g/cm^3 because the energy response correction factors r_w provided by the supplier were only given for these two densities.

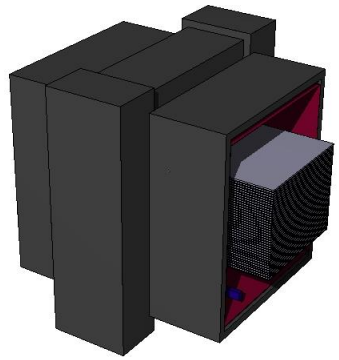

Figure 2a – Model of the TGMC system

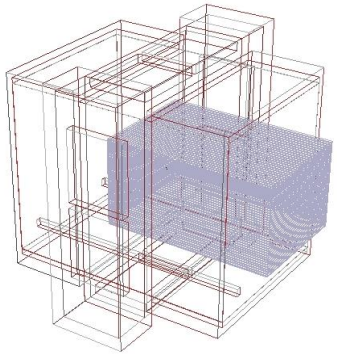

Figure 2b – Structure of the model

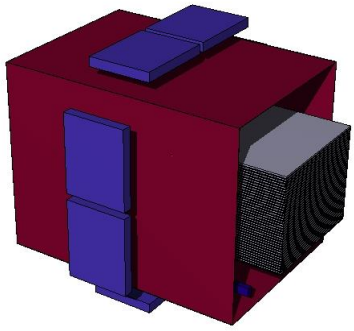

Figure 2c – Detail of the model: d detectors in purple

Determination of the energy threshold in the model

By the Monte Carlo simulations it is possible to determine, for each photon emitted by the source and for each detector, the number of counts for each 2 keV energy interval. Since a variation of the high voltage applied to the photomultiplier implies a variation of the energy threshold of the detector, in order to properly use the simulations produced, it is necessary to determine the value of this threshold in the model used.

For this purpose, the model has been created based on the simplest and most reproducible source-detector configuration which consists in a point source of ^{60}Co placed in the calibration point located inside the measurement tunnel. For each detector, the threshold is calculated as the energy for which is minimum the difference between the detector response experimentally determined and the one determined by the simulation.

The energy threshold for each d detector at the optimal high voltage is reported in Table 1.

Detector	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**
E_s [keV]	318	302	330	272	290	250	252	194

Table 1 – Threshold energy for each d detector at the optimal high voltage.

Validation of the model

The model validation for the TGMC is performed by comparing the response of the system for a homogeneous matrix source of ^{40}K with the results of the corresponding simulated geometry. The specific

activity of the matrix has been determined *a priori* by the JRC Ispra accredited Laboratory [13]. ^{40}K as potassium nitrate powder was chosen for its easy availability in huge volumes, volume fitting capability as well it emits a mono-energetic photon comparable to the ones of ^{60}Co.

Table 2 shows the deviation of the simulation results using the model from the actual TGMC measurement. The difference is approximately 12% and therefore the model is considered a good approximation of the actual system.

TGMC	**Response [cps]**	**Δ[%]**
Measurement	2372,455	/
Simulation	2659,771	**+12,11**

Table 2 – Monte Carlo model validation

Use of the model for the verification of the global transfer function

Although the actual calibration is carried out considering the responses of the d detector for three positions of the container into the tunnel, the simulations for the determination of f^d_T were carried out only in the configuration with the container placed in the centre of the tunnel. This choice is justified by the fact that also the energy response correction factors r_w were determined by the supplier only in this configuration.

The average deviation of the correction factors f_T determined by Monte Carlo for a heterogeneous calibration standard container and a homogeneous ideal container is reported in Table 3. The choice of using the average deviation in this case is justified by the fact that the energy response correction factors r_w are determined by the supplier as a mean value on the eight detectors. Anyway, it should be noticed that the calibration carried out with the standard container is overestimating in any case the correction factor f_T.

Density 0.5 g/cm^3			**Density 2.1 g/cm^3**		
$f_{T\ -\ Heterogeneous}$	$f_{T\ -Homogeneous}$	*Δ[%]*	$f_{T\ -\ Heterogeneous}$	$f_{T\ -Homogeneous}$	*Δ[%]*
155,0467925	135,1199731	+14,75	459,7014325	334,1076519	+37,21

Table 3 - Average deviations of the correction factors for the transmission of the photons in the source f^d_T

Once the η_i values of the function η have been calculated by Monte Carlo for each of the seven reference energies E_i, both for the simulations with standard (heterogeneous) container and for those with ideal homogeneous container, it is possible to determine the curves $\eta(E)$ by means of least squares regression (see Figures 3 and 4).

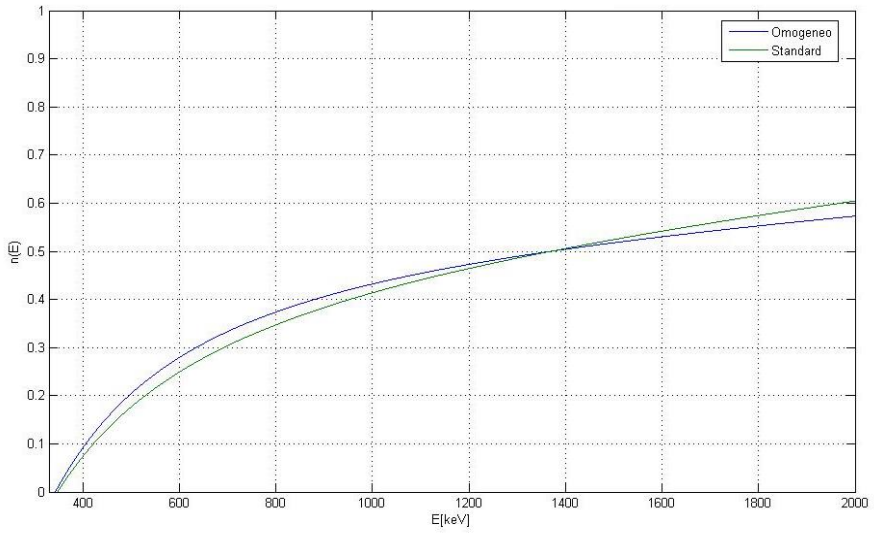

Figure 3 – Curves η(E) at low density: 0.5 g/cm^3.

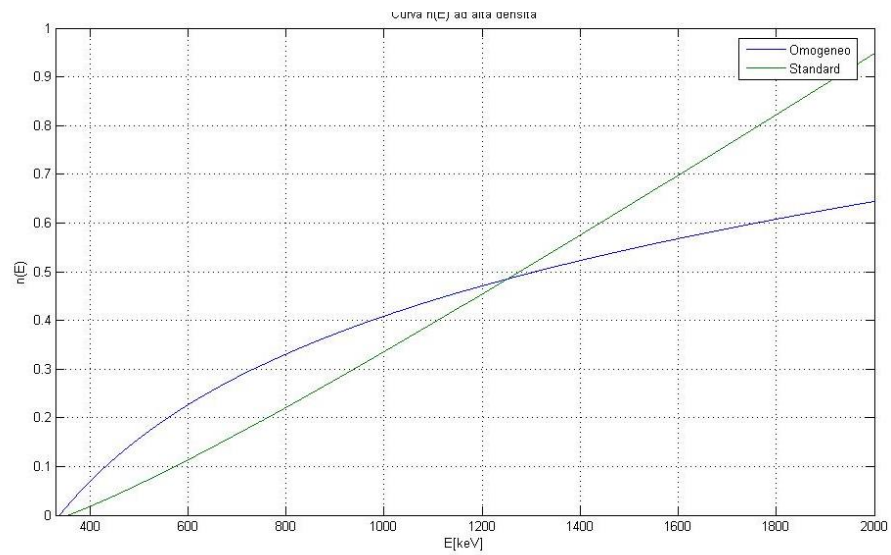

Figure 4 – Curves η (E) at high density: 2.1 g/cm^3.

It is then possible to determine the deviation curves at different energies as follows:

$$\Delta(E) = \frac{\eta_{inhomogeneous} - \eta_{homogeneus}}{\eta_{homogeneous}} \tag{7}$$

It can be observed that the calibration performed with the calibration standard container is overestimating the homogeneous one only at energies below 1250 keV where the correction factor r_w is underestimated. At higher energies the calibration performed with the calibration standard container overestimates r_w resulting in underestimation of the activity in case of perfect homogeneity. The deviation curves at different energies are shown hereinafter (see Figures 5 and 6).

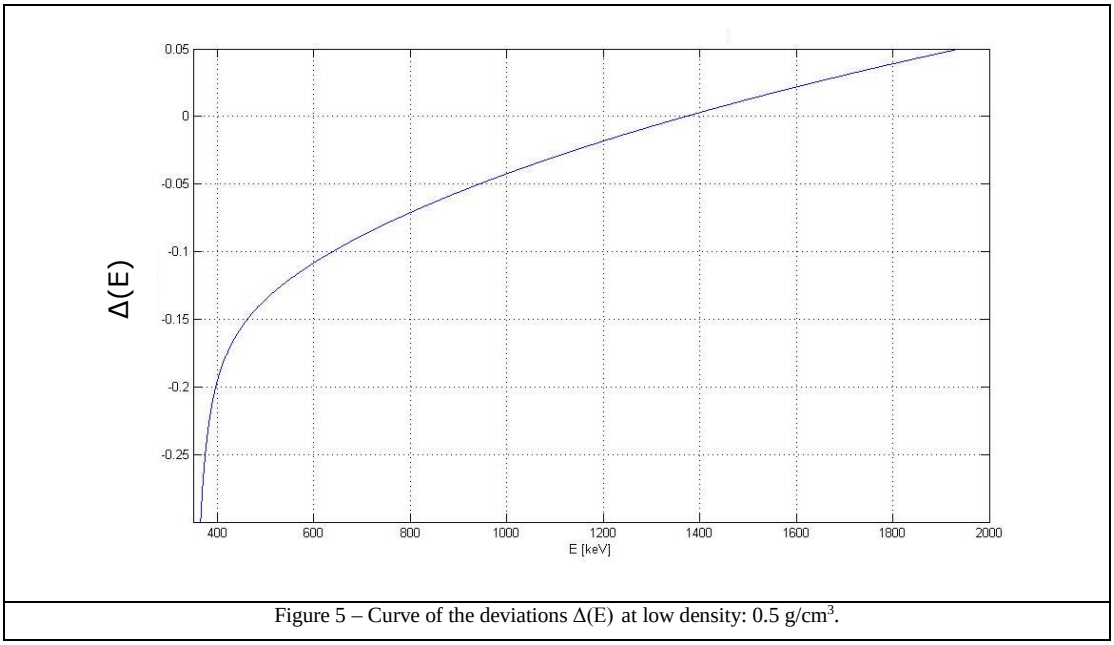

Figure 5 – Curve of the deviations Δ(E) at low density: 0.5 g/cm^3.

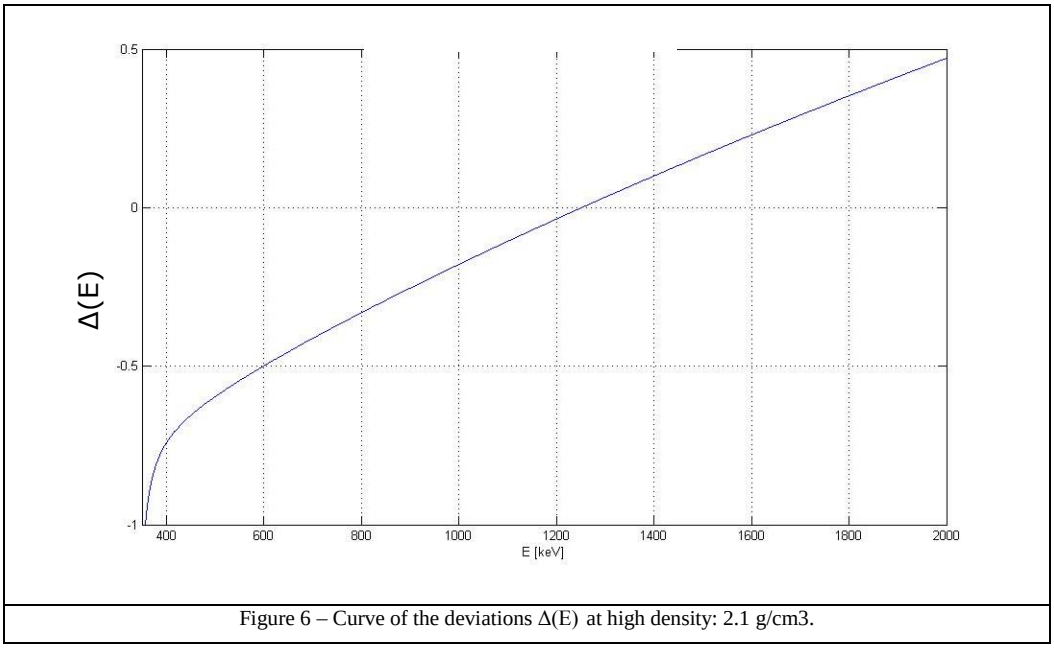

Figure 6 – Curve of the deviations Δ(E) at high density: 2.1 g/cm3.

The deviations of the energy response correction factors r_w determined by the Monte Carlo model of a standard container from the corresponding values determined with the model of an ideal container with a density of 0.5 g/cm^3 and of 2.1 g/cm^3 and for the two radionuclides ^{137}Cs (662 keV) and ^{88}Y (1836 keV), are reported in Table 4. The two nuclides have been selected out of the seven used for the r_w calculation because of their representativeness of the energy range of interest.

Configuration	**Density 0.5 g/cm^3**		**Density 2.1 g/cm^3**	
	662 keV	1836 keV	662 keV	1836 keV
Deviation [%]	-9,56	+4,18	-4,46	+37,35

Table 4 – Deviation of the energy response correction factors r_w

Once the deviations of the single correction factors f_T and r_w have been determined it is possible to calculate the deviation of the overall transfer function from its true value. The calibration performed by the calibration standard container is overestimating the fully homogeneous one only at low energies (below 1250 keV).. The deviation of the overall transfer function from its true value is shown in Table 5.

Configuration	**Density 0.5 g/cm^3**		**Density 2.1 g/cm^3**	
	662 keV	1836 keV	662 keV	1836 keV
Deviation [%]	+11,20	-2,75	+8,50	-34,93

Table 5 – Deviation of the overall transfer function from its true value.

6. Conclusions

The simulations showed a deviation of the global transfer function determined by the calibration container from the one determined in pure homogeneous conditions. Consequently the activity calculated by the system as described by Eq. (1) may underestimate the actual activity. Therefore, a safety factor f_{mon} equal to 1.25 has been introduced in the algorithm of the TGMC for the calculation of the activity giving the following:

$$A_W^d = \left(\dot{R}_\Delta^d + z_{sta}^d\right)\frac{f_T^d \cdot f_{mon}}{r_W} \tag{8}$$

The safety factor has been calculated by linear interpolation between the two densities considered for the maximum density of the JRC containers allowed by the Italian Authorities (about 1.6 g/cm^3 corresponding to 1000kg for a 0.65m^3 containers) at the maximum energy considered (1836 keV).

Similarly to z^d_{sta}, the safety factor f_{mon} increases the value of the measured activity in such a way that, for any measurement condition, it will never be less than the true value because of a possible in-homogeneity (matrix and source) of the containers.

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