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A new approach in gamma-ray scanning of rotating drums containing radioactive waste

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ABSTRACT

The paper presents a new approach in the integral gamma scanning of rotating waste drums based on solving a first-kind Fredholm integral equation using a small number of measurements. Both numerical experiments and experimental results show that the nuclide inventory of waste drums can be determined accurately using the new approach especially when the waste drums contain multi-gamma emitters.

1. Introduction

Integral gamma scanning method is routinely used to characterize the nuclide inventory of radioactive waste packages because it is simple, fast and cheap (IAEA, 2007). The classical integral gamma scanning method involves only one detector and the waste drum is usually rotated continuously during the spectrum acquisition. A single measurement is performed and the peak efficiency is calculated on the basis of the simplifying assumption of waste homogeneity. This assumption may lead to large errors of measurement (Bronson et al., 2008; Rottner, 2007).

The paper presents a new approach in the integral gamma scanning of rotating waste drums based on solving a first-kind Fredholm integral equation using a small number of measurements. It aims at extending the applicability of the integral gamma scanning method to the measurement of waste drums with relatively homogeneous matrix and heterogeneous activity distribution at the expense of performing several measurements. A model for computing the peak efficiency was developed to discretize the first-kind Fredholm integral equation and obtain an ill-conditioned system of equations which was solved using the Tikhonov regularization with non-negativity constraint.

The performance of the approach was firstly investigated using synthetic data. Numerical experiments show that accurate results can be obtained in the measurement of waste drums with relatively homogeneous matrix and heterogeneous activity distributions especially for nuclides emitting two or more gamma rays. Also, numerical experiments show that reasonable results can be obtained for multi-gamma emitters even if the mass distributions are highly heterogeneous.

The approach was applied to the activity measurement of a certified linear ^{152}Eu source placed at different radial positions into a 220-Litre drum filled with cement. Accurate results were obtained using only few measurements for each position of the source.

2. Theoretical basis

2.1. Computational model of the peak efficiency for uncollimated HPGe detectors

Fig. 1 shows the geometry used for integral gamma scanning of rotating waste drums. In this geometry, the axis of the rotating waste drum is perpendicular to the detector axis. Using an uncollimated HPGe detector placed at the distance d from the drum axis and height z_0, the peak counting rate (corrected for dead time effects and background), $R_p(E,\mu,d,z_0)$, due to photons of energy E, is given by (Stanga et al., 2010)

$$R_p(E,\mu,d,z_0) = \int_0^R \int_0^h \varepsilon_c(E,\mu,r,z,d,z_0)\Lambda_\theta(r,z)r\,dr\,dz \quad (1)$$

where

$$\varepsilon_c(E,\mu,r,z,d,z_0) = \frac{\varepsilon_{ref}(E)}{2\pi}\int_0^{2\pi}\left(\frac{L_{ref}}{L(\theta)}\right)^2 f_{ang}(\theta)\exp\left[-\mu(\theta)t(\theta)-\mu_w t_w(\theta)\right]d\theta \quad (2)$$

and

$$\Lambda_\theta(r,z) = \int_0^{2\pi}\Lambda_V(r,z,\theta)d\theta \quad (3)$$

here, $\varepsilon_c(E,\mu,r,z,d,z_0)$ represents the peak efficiency of a point source in rotation characterized by the coordinates r and z, $\Lambda_V(r,z,\theta)$ is the volume activity, $\varepsilon_{ref}(E)$ is the peak efficiency of the reference source located on the detector axis at the distance

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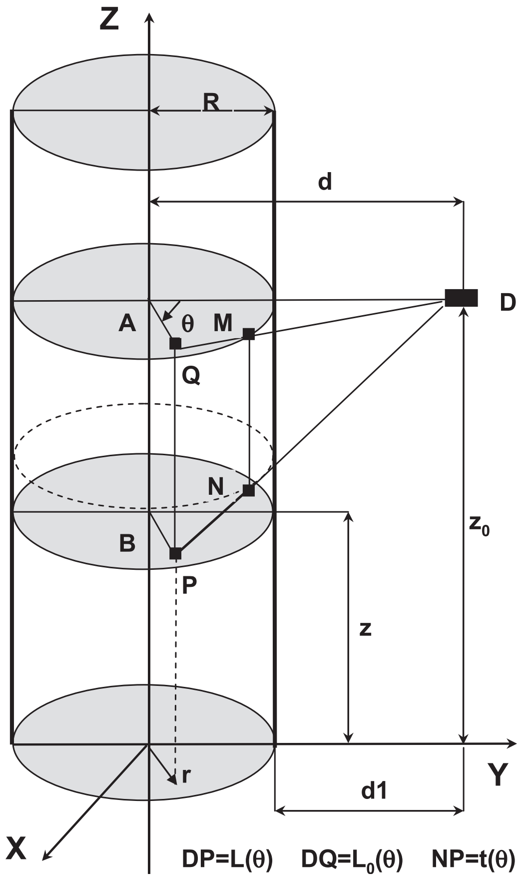

Fig. 1. Schematic drawing of the waste drum scanning system with the drum located between the extreme shaded areas and the HPGe-detector (D) located at the distance *d* from the drum axis.

L_{ref}, $L(\theta)=\sqrt{L_0^2+(z-z_0)^2}$, $L_0(\theta)=\sqrt{r^2+d^2-2rd\cos\theta}$, $f_{ang}(\theta)$ is the angular correction factor which characterizes the angular response of the detector, $\mu(\theta)$ is the average value of the linear attenuation coefficient over the attenuation length $t(\theta)=(\sqrt{R^2-[(dr/L_0)\sin\theta]^2}$ $-(r/L_0)(d\cos\theta-r))(L(\theta)/L_0)$ in the waste matrix, μ_w and $t_w(\theta)=\sqrt{(R+w_t)^2-[(dr/L_0)\sin\theta]^2}-\sqrt{R^2-[(dr/L_0)\sin\theta]^2}$ are the linear attenuation coefficient and the attenuation length in the wall of the drum (w_t-thickness of the drum wall). Taking $\mu(\theta)=\mu_0+\delta(\theta)$ and applying the mean value theorem, Eq. (2) becomes

$$\varepsilon_c(E,\mu,r,z,d,z_0)$$
$$=\frac{\varepsilon_{ref}}{2\pi}\alpha(r,z,d,z_0)\int_0^{2\pi}\left(\frac{L_{ref}}{L(\theta)}\right)^2 f_{ang}(E)\exp\left[-\mu_0 t(\theta)-\mu_w t_w(\theta)\right]d\theta \tag{4}$$

where μ_0 is the volume-average value of the linear attenuation coefficient of the waste matrix, $\alpha(r,z,d,z_0)=\exp(-\delta t)$ is the correction factor for the matrix heterogeneity, $\delta=\delta(\theta_0)$, $t=t(\theta_0)$ and $\theta_0\in(0,2\pi)$. For rotating drums with homogeneous matrix, α is equal to unity. When the degree of heterogeneity of the matrix is small, α is close to unity.

2.2. Description of the new approach

The rotating waste drum is measured for different positions of the detector defined by $d(k)$ and $z_0(l)$, where $k=1,2,\ldots k_{max}$ and $l=1,2,\ldots l_{max}$. In this way, the peak count rates $R_{peak}(i)$ corresponding to the ith position of the detector $(i=(l-1)k_{max}+k)$ are obtained. Here we consider that the maximum number of measurements is twenty and accurate measurement results are used.

To obtain a computer approximation of the activity, the integral Eq. (1) is discretized by dividing the drum volume into a system of voxels of equal volumes composed of cylindrical rings and central cylinders. As a result, integral Eq. (1) is converted into the following system of algebraic equations

$$R_{peak}(i)=\sum_{j=1}^{N}\varepsilon_c(i,j)\alpha(i,j)\Lambda(j)\ \mathrm{i}=1,2,\ldots\mathrm{M} \tag{5}$$

where $\Lambda(j)$ is the activity of the jth rotating point source located in the jth voxel and characterized by the coordinates $r(p)=R\sqrt{(2p-1)/(2p_{max})}$ and $z(m)=h(2m-1)/(2m_{max})$, $(p=1, 2,\ldots p_{max}$, $m=1,2,\ \ldots m_{max}$, $j=(m-1)p_{max}+p$, $N=p_{max}m_{max})$, $\varepsilon_c(i,j)$ and $\alpha(i,j)$ are, respectively, the system matrix (peak efficiency of the detector) and the correction factor for the ith position of the detector and the jth rotating point source. To compute $\varepsilon_c(i,j)$, values of $\varepsilon_{ref}(E)$ for different gamma energies were taken from the calibration certificate of the detector manufacturer ($L_{ref}=30$ cm) and these values have relative standard uncertainties of 3%.

For matrices with a low degree of heterogeneity we can assume that $\alpha(i,j)$ depends weakly on the detector position and we have $\alpha(i,j)\approx\alpha(1,j)=\alpha(j)$. In this case, Eq. (5) becomes

$$R_{peak}(i)=\sum_{j=1}^{N}\varepsilon_c(i,j)\Lambda_a(j) \tag{6}$$

where $\Lambda_a(j)=\alpha(j)\Lambda(j)$. For homogeneous matrices $\alpha(j)=1$ $(j=1,2,\ldots\mathrm{N})$ and the total activity, for a given nuclide, is $\Lambda_T=\sum_{j=1}^{N}\Lambda(j)$. For matrices with a low degree of heterogeneity, we have

$$\Lambda_T=\sum_{j=1}^{N}\Lambda_a(j)/\sum_{j=1}^{N}\alpha(j)c(j)=\Lambda_T^{(a)}/fc \tag{7}$$

where $c(j)=\Lambda(j)/\Lambda_T$, $\Lambda_T^{(a)}=\sum_{j=1}^{N}\Lambda_a(j)$ and $fc=\sum_{j=1}^{N}\alpha(j)c(j)$. The correction factors, $\alpha(j)$, take values in a narrow interval around unity due to the low degree of heterogeneity of the matrix. Because *fc* is the weighted mean of the correction factors, we have $fc\cong 1$ and $\Lambda_T\cong\Lambda_T^{(a)}$. In this way, it was proved that the new approach can also be applied to the measurement of waste drums with relatively homogeneous matrix because the additional error due to the matrix heterogeneity is small.

To compute Λ_T, the system of algebraic Eq. (6) is firstly normalized in such a way that the Euclidean norms of the matrix and the right-hand side are equal to unity. As a result, we obtain, in matrix form, the following system of equations

$$Ax=b \tag{8}$$

where $A=\frac{\varepsilon_c}{\|\varepsilon_c\|}$, $b=\frac{R_{peak}}{\|R_{peak}\|}$, $x=\frac{\|\varepsilon_c\|}{\|R_{peak}\|}\Lambda_{approx}$, $\|\varepsilon_c\|$ and $\|R_{peak}\|$ are Euclidean norms.

To solve the ill-conditioned system of equations (8), many regularization methods are available. We choose the Tikhonov regularization method with non-negativity constraint because it is fast and simple. The idea of the (zeroth-order) Tikhonov regularization is to minimize the functional $J(x)=\|Ax-b\|^2+\alpha\|x\|^2$ where α (regularization parameter) is a real positive number (Tikhonov, 1963a, 1963b). The minimization problem is equivalent to the equation

$$(AA^T+\alpha I)x=A^Tb \tag{9}$$

where I is the identity matrix and A^T is the transpose of A. To obtain a non-negative solution for Eq. (9), the non-negative least squares algorithm introduced by Lawson and Hanson (1974) was applied. The non-negativity constraint is physically realistic because the components, $\Lambda(j)$, of the activity distribution cannot be negative.

The choice of the regularization parameter values is important. Several methods are available for choosing α, such as the *L*-curve method (Hansen, 1992) but they need a considerable amount of computation. We selected a fixed value for α ($\alpha = 2.5 \times 10^{-7}$) which was used in all calculations. Selecting a fixed value for α is the simplest method for choosing the regularization parameter but it is not optimal.

The approach described above was implemented in MATLAB and several codes were developed for computing Λ_T in different numerical experiments.

3. Numerical experiments

3.1. Synthetic data

The performance of the new approach in the measurement of Λ_T for 220-Litre waste drums was firstly tested using synthetic data. These data were obtained by taking $\Lambda_T = 10$ MBq and considering different types of mass and activity distributions. Characteristics of these distributions are shown in Table 1. We produced the synthetic data, $R_{peak}^{(s)}$, using both the GESPECOR software and a simpler forward model (point detector model) than that described in Section 2.1 (Sima et al., 2001, Notea, 1971). A simple forward model taking into account the mass distribution was also used to produce synthetic data for MD2 distribution.

The stability of the approach at different noise levels was tested using perturbed counting rates. Thus, approximating the Poissonian distribution of R_{peak} with a Gaussian distribution, we generated perturbed counting rates as

$$R_{peak} = R_{peak}^{(s)} + \sqrt{R_{peak}^{(s)}/T} \otimes randn(M,1) \tag{10}$$

where T is the counting time, randn(M,1) is a random vector from a normal distribution with zero mean and unit standard deviation and the symbol $\otimes$ denotes element-wise multiplication. The noise level for the vector b is given by the noise-to-signal ratio $NRS = \|R_{peak}^{(s)}/T\| / \|R_{peak}\|$. We generated noisy data 1000 times for a given activity distribution and a given value of *NRS*. As a result, we obtained 1000 solutions for Eq. (8) and 1000 values for Λ_T. From the distribution of Λ_T values, we calculated the standard deviation of Λ_T which quantifies the stability of the approach.

The accuracy of the approach depends both on its stability to different noise levels (propagated noise error) and regularization error. It was investigated for a set of 220-Litre waste drums containing AD4 activity distributions. For given values of the waste density and *NRS*, we generated 1000 different activity distributions type AD4 and the corresponding noisy data. In this way, we obtained 1000 solutions for Eq. (8) and 1000 values for Λ_T. From the distribution of Λ_T values, we calculated the average value of Λ_T, the associated standard deviation and the 95% coverage interval. The reason for using AD4 is based on the fact that a typical waste drum is likely to contain at least 3 equivalent point sources randomly distributed in the drum volume rather than a single localized source (Venkataraman et al., 2005).

3.2. Application of the approach to waste drums with homogeneous matrix using synthetic data

The approach was applied to the measurement of 220-Litre waste drums with homogeneous matrix containing ^{137}Cs or ^{60}Co which are key nuclides in radioactive waste assays. The integral Eq. (1) was discretized using $N = 7 \times 7 = 49$ voxels. Values for the components of the vector R_{peak} corresponding to energies of interest (662 keV—^{137}Cs; 1173 and 1332—^{60}Co) were calculated for $d1 = 75,100,125,150$ cm ($k_{max} = 4$) and $z_0 = 22,33,44,55,66$ cm ($l_{max} = 5$), where $d1 = d - R$ is the distance between the detector and the drum wall.

Results regarding the stability of the approach are shown in Table 2. In this table, calculated values of the standard deviation of Λ_T, corresponding to ^{137}Cs and ^{60}Co, are given for single, duplicate and triplicate measurements. For each value of *NRS*, the standard deviation was calculated for three values of the waste density and three types of activity distributions (AD1, AD2 and AD3). As one can see from Table 2, the stability of the approach depends not only on the noise level, the waste density and the type of activity distribution but also on the nuclide. Thus, the stability of the approach is better in the case of ^{60}Co than in the case of ^{137}Cs. From numerical tests we found that the stability depends both on the number of gamma rays emitted by the nuclide and the difference between their energies. A very good stability is obtained for multi-gamma emitters with big differences between energies. Also, Table 2 shows that small values for the standard deviation of Λ_T are obtained only for low values of the waste density and *NRS*. Using duplicate or triplicate measurements, the stability is significantly improved. A good stability is obtained for AD2 activity distribution but the regularization error is large in this case.

Results regarding the accuracy of the approach for a set of 1000 waste drums with homogeneous matrix containing AD4 different activity distributions are shown in Table 3. In this table, the average value of Λ_T, the associated standard deviation and the 95% coverage interval, corresponding to ^{137}Cs and ^{60}Co, are given for different values of the waste density and *NRS*. In Table 3 are also shown, for comparison, the results obtained with the classical method (integral gamma scanning method based on the assumption of homogeneity) using the same activity distributions. We can see from Table 3 that the accuracy of the approach corresponding to ^{60}Co is better than that corresponding to ^{137}Cs, as expected. Both for ^{137}Cs and ^{60}Co, the approach is satisfactory because the average value of Λ_T is close to the true value $\Lambda_T = 10$ MBq (see Section 3.1). Hence, in contrast with the classical method, the new approach does not overestimate significantly the overall activity in a set of waste drums containing randomly distributed sources (Rottner, 2007). Also, the new approach has smaller standard deviations and narrower coverage intervals than the classical method in the case of ^{60}Co. The same results were obtained for ^{137}Cs but only for $NRS = 0.01$. However, standard

Table 1
Activity and mass distributions used for obtaining synthetic data.

Activity and mass distributions	Characteristics
AD1	Homogeneous activity distribution contained in the whole drum volume $V = \{(r,z,\theta)$: $r \in (0,28.5$ cm$)$, $z \in (0,88$ cm$)$, $\theta \in (0,2\pi)\}$
AD2	Homogeneous activity distribution contained in the volume $V_1 = \{(r,z,\theta)$: $r \in (0,7.5$ cm$)$, $z \in (0,88$ cm$)$, $\theta \in (0,2\pi)\}$
AD3	Homogeneous activity distribution contained in the volume $V_2 = \{(r,z,\theta)$: $r \in (21,28.5$ cm$)$, $z \in (0,88$ cm$)$, $\theta \in (0,2\pi)\}$
AD4	Three point sources randomly distributed in the whole drum volume $V = \{(r,z,\theta)$: $r \in (0,28.5$ cm$)$, $z \in (0,88$ cm$)$, $\theta \in (0,2\pi)\}$
MD1	Homogeneous matrix in the whole drum volume
MD2	Homogeneous matrix of density ρ_1 in the volume $V_3 = \{(r,z,\theta)$: $r \in (0,20$ cm$)$, $z \in (0,88$ cm$)$, $\theta \in (0,2\pi)\}$ Homogeneous matrix of density ρ_2 ($\rho_1 \neq \rho_2$) in the volume $V_4 = \{(r,z,\theta)$: $r \in (20,28.5$ cm$)$, $z \in (0,88$ cm$)$, $\theta \in (0,2\pi)\}$

deviations smaller than 10% and narrow coverage intervals are obtained only for low values of the waste density and *NRS* especially in the case of ^{137}Cs. Using duplicate or triplicate measurements, better results can be obtained.

3.3. *Application of the approach to waste drums with heterogeneous matrix using synthetic data*

The new approach was applied to the measurement of a set of 1000 waste drums (220-Litre) with heterogeneous matrix (MD2 mass distribution) containing ^{60}Co. The assessment of the accuracy was performed in the same conditions as those described above for waste drums with homogeneous matrix. Results regarding the accuracy of the approach in the measurement of rotating waste drums with four different MD2 mass distributions are shown in Table 4. We can see from Table 4 that reasonable results were obtained even if the mass distributions are highly heterogeneous. For mass distributions with low degree of heterogeneity (ρ_1=0.8 g/cm^3, ρ_2=1.2 g/cm^3 and ρ_1=1.2 g/cm^3, ρ_2=0.8 g/cm^3), the results are close to those obtained for corresponding homogeneous matrices (ρ=1 g/cm^3). Hence, as it was shown previously (see Section 2.2), the approach can also be applied to the assay of waste drums containing matrices with low degree of heterogeneity.

Table 2
Results regarding the stability of the approach (for single, duplicate and triplicate measurements) applied to the measurement of waste drums with homogeneous matrix containing activity distributions of ^{137}Cs or ^{60}Co.

Activity distribution	*NRS*	Relative standard deviation (%)								
		ρ_w (g/cm^3)								
		Single measurements			Duplicate measurements			Triplicate measurements		
		0.5	1.0	2.0	0.5	1.0	2.0	0.5	1.0	2.0
^{137}Cs										
AD1	0.01	11.0	17.0	27.0	8.0	12.4	18.8	3.5	5.7	10.6
	0.03	18.0	36.0	54.0	12.8	25.7	41.5	8.2	14.9	21.8
AD2	0.01	6.3	7.6	6.8	4.8	5.4	5.9	3.9	4.6	3.8
	0.03	14.0	19.0	22.0	9.6	13.6	16.0	7.8	10.8	13.1
AD3	0.01	11.7	21.7	33.2	8.5	15.0	22.9	7.0	12.5	19.5
	0.03	18.8	37.5	59.6	13.2	27.8	42.1	10.8	22.4	34.3
^{60}Co										
AD1	0.01	3.6	4.0	10.8	2.9	2.7	8.3	2.02	2.3	6.5
	0.03	9.2	10.8	19.1	5.8	7.7	13.8	5.2	6.09	11.4
	0.05	11.9	17.0	26.4	8.3	12.4	18.9	6.7	9.9	15.1
AD2	0.01	1.6	0.9	0.5	1.2	0.6	0.35	1.0	0.5	0.3
	0.03	5.5	4.5	2.6	3.4	3.2	1.8	3.3	2.4	1.5
	0.05	8.4	7.6	5.5	5.6	5.5	4.1	5.0	4.5	3.2
AD3	0.01	4.4	5.6	12.4	3.0	3.8	9.0	2.3	3.3	7.3
	0.03	9.2	14.4	24.9	6.5	9.7	18.5	5.3	8.4	14.8
	0.05	12.5	20.5	37.3	8.5	14.4	25.6	6.9	11.9	20.2

4. Experimental results

The performance of the new approach was tested experimentally by measuring a 220-Litre calibration drum filled with Portland cement and containing a linear ^{152}Eu source. The calibration drum is provided with 7 hollow vertical tubes placed at different radial positions. The certified linear ^{152}Eu source of length 62 cm was inserted into vertical tubes and the drum

Table 4
Results regarding the accuracy of the approach for a set of waste drums with heterogeneous matrix containing AD4 activity distributions of ^{60}Co.

MD2 characteristics (g/cm^3)	*NRS*	Average Λ_T [a] (MBq)	Relative standard deviation (%)	95% coverage interval (MBq)
ρ_1=0.2	0.01	9.75	13.0	[7.8,12.4]
ρ_2=1.8	0.03	10.0	17.4	[6.8,13.1]
ρ_1=0.8	0.01	9.97	6.00	[8.9,11.1]
ρ_2=1.2	0.03	10.1	13.0	[7.7,12.6]
ρ_1=1.8	0.01	10.9	7.50	[9.3,12.4]
ρ_2=0.2	0.03	11.0	12.9	[8.0,13.4]
ρ_1=1.2	0.01	10.3	4.64	[9.3,11.2]
ρ_2=0.8	0.03	10.4	11.7	[8.0,12.8]

[a] The true value of Λ_T is 10 MBq.

Table 3
Results regarding the accuracy of the approach and the classical method for a set of waste drums with homogeneous matrix containing AD4 activity distributions of ^{137}Cs and ^{60}Co.

Waste density (g/cm^3)	*NRS*	New approach			Classical method		
		^{137}Cs			^{137}Cs		
		Average Λ_T[a] (MBq)	Relative standard deviation (%)	95% coverage interval (MBq)	Average Λ_T[a] (MBq)	Relative standard deviation (%)	95% coverage interval (MBq)
0.5	0.01	9.88	10.6	[8.0,12.1]	11.1	12.9	[8.5,13.9]
	0.03	9.90	18.6	[6.6,13.4]			
1.0	0.01	10.2	19.7	[6.2,14.1]	12.8	30.4	[5.9,20.4]
	0.03	10.9	37.4	[4.5,19.3]			
2.0	0.01	10.2	34.3	[4.8,17.8]	14.8	58.3	[1.5,31.0]
	0.03	12.2	62.0	[1.8,22.6]			
		^{60}Co			^{60}Co		
0.5	0.01	10.0	3.64	[9.3,10.7]	10.7	8.7	[9.1,12.6]
	0.03	9.92	8.70	[8.4,11.6]			
	0.05	9.94	11.7	[7.8,12.2]			
1.0	0.01	10.1	4.80	[9.1,11.0]	11.8	21.4	[7.4,16.6]
	0.03	10.2	12.0	[7.8,12.6]			
	0.05	10.3	18.3	[6.8,14.1]			
2.0	0.01	10.4	13.1	[7.9,13.4]	13.5	44.9	[3.8,24.9]
	0.03	11.0	21.9	[6.3,15.4]			
	0.05	11.5	31.4	[5.5,18.3]			

[a] The true value of Λ_T is 10 MBq.

Table 5
Peak counting rates obtained in the measurement of the 220-L calibration drum containing ^{152}Eu measurement results obtained using both the new approach and the classical method.

Source position (cm)	$d1$ (cm)	Peak counting rate (s^{-1})						Measurement results			
		z_0=30 cm		z_0=41 cm		z_0=52 cm		New approach		Classical method	
		Energy (keV)						Λ_T (MBq)	Relative difference[b] (%)	Λ_T (MBq)	Relative difference (%)
		964	1408	964	1408	964	1408				
rs[a]=24.5	125	23.58	29.20	23.60	29.37	23.21	29.14	52.9	1.7	92.3	77.5
	150	18.80	23.19	18.65	23.01	18.49	23.19				
rs=22	100	22.74	30.31	23.36	30.24	23.03	30.31	50.3	3.2	64.9	24.9
	125	17.11	23.32	17.80	23.27	18.10	23.42				
rs=16	100	10.27	16.10	10.71	16.24	9.96	16.15	56.5	8.7	28.4	45.4
	125	7.78	11.89	7.78	12.05	7.75	12.00				
rs=11	75	8.89	16.39	8.93	16.38	9.48	16.54	58.6	12.7	15.7	69.7
	100	6.34	11.27	6.17	11.20	6.61	11.19				
rs=0	75	5.40	10.58	5.36	10.73	5.03	10.52	44.1	15.2	9.1	82.4
	100	3.52	7.19	3.58	7.12	3.56	7.10				

[a] rs Is the distance between the ^{152}Eu source and the drum axis.
[b] Relative difference=$(\Lambda_T-\Lambda_{cert})/\Lambda_{cert}$ and Λ_{cert}=52.2 MBq.

rotated during the measurements. The activity of the source was equal to 5.22 MBq (1.06.2010) with a relative standard uncertainty of 2.5%. The density of the cement was determined by weighting and a value of 2.1 g/cm^3 was obtained.

An ISO-CART gamma spectrometric system was used in measurements. It contains a SMART-1 HPGe detector type GEM 25P4, which is connected to a digiDART multichannel analyzer controlled by a laptop computer. ISOTOPIC 3.1 software was used for data acquisition and spectrum analysis. In all measurements the average dead time loss was smaller than 20% and counting times were chosen to achieve counting statistics better than 3%.

Table 5 shows the values of the peak counting rate corresponding to two gamma lines of ^{152}Eu determined for different detector and source positions. In the same table are also shown the measurement results obtained using both the new approach and the classical method. To compute the total activity Λ_T, the integral Eq. (1) was discretized using $N=7\times 7=49$ voxels and the values for the components of the vector R_{peak} were taken from Table 5. One can see from Table 5 that the new approach gives better results than the classical method at the expense of performing six measurements. Thus, the new approach gives relative differences between the true and calculated values of Λ_T smaller than 16% while the classical method has relative differences of up to 82%.

5. Conclusions

This paper presents a new approach in the integral gamma scanning of rotating waste drums. The performance of the approach was tested using both synthetic and real data. Results show that the approach is able to determine accurately the total activity of a given radionuclide from rotating waste drums with relatively homogeneous matrix and heterogeneous activity distribution especially for multi-gamma emitters. Also, numerical experiments show that reasonable results can be obtained for multi-gamma emitters even if the mass distributions are highly heterogeneous.

Experimental results show that the approach can be applied to the measurement of high density waste drums (ρ=2.1 g/cm^3) containing multi-gamma emitters. Thus, relative differences between the true and calculated values of Λ_T smaller than 16% were obtained in the measurement of a certified linear ^{152}Eu source placed at different radial positions (including the central position) into a 220-Litre calibration drum filled with cement.

In conclusion, promising results were obtained with the new approach especially for multi-gamma emitters. Further research is necessary to (1) find a strategy for choosing the optimal regularization parameter and (2) extend the applicability of the approach to the measurement of waste drums with highly heterogeneous matrix using both collimated and uncollimated detectors.

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