

Study on Effect of Density Heterogeneity for Nuclear Waste Drum Samples in Segmented Gamma Scanning Analysis

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Abstract With a segmented gamma scanning (SGS) equipment, the non-destructive transmission and emission measurement are experimented for eight radioactive waste samples with heterogeneous activity and matrix distribution. Radioactivity of both ^{137}Cs and ^{60}Co sources in those samples is calculated through a self-absorption correction method. The results show that the relative deviations between corrected and actual radioactivity of eight samples are all less than 5%. Then the effect of density heterogeneity for nuclear waste drum samples was studied, indicating that the SGS analyzed radioactivity will be deviated from its actual radioactivity along with different density of samples. With filling material density heterogeneity and average density of samples increasing, the relative deviations between corrected and actual radioactivity trend to increase gradually. The heterogeneity immediately determines the scale of relative deviations in SGS analysis. The density of samples, the energy and transmission ability of gamma ray are main factors of the deviation in SGS analysis.

Keywords Segmented gamma scanning · Radioactive waste · Density heterogeneity · Linear attenuation coefficient · Self-Absorption correction

1 Introduction

The segmented gamma scanning (SGS) technique is being widely employed to perform non-destructive assay of the radioactive waste drum. The SGS technique can quickly identify the kind of radioactive nuclide and the radioactivity of the drum. In SGS, a radioactive waste drum is rotated in front of a collimated detector. At each segment, the detector takes a spectrum. After all segments of the drum are

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detected, the radioactivity of the drum is calculated. In the early 1970s, Parker et al. [1] of Los Alamos National Laboratory (LANL) had begun to research the SGS technique and Espartero [2] applied it into the field of the low radioactive waste measurement. In the 1990s, Lu [3], Zhu [4], Cao [5] and some other Chinese researchers began to research the SGS technique. With the self-correction factors obtained by SGS experimental measurement, the contents of the uranium and the plutonium were analyzed in radioactive residues and wastes based on the SGS technique. Xu [6] has designed a basic model of standard source for the radioactive waste drum formed with rotating the waste drum in which several linear sources were put. Combined the Monte Carlo simulation with the experimental measurement, the effect of radial heterogeneity in SGS analysis was studied by Wang [7].

The deviation of SGS analysis consists of deviation about radial or axial heterogeneity of samples and axial deviation caused by the stratified strategy. It could reduce the deviation of analysis and improve the accuracy for a particular SGS system that adopted the suitable stratified strategy particularly [8]. This work discusses the problems for different density heterogeneity on a specific sample segment.

Based on the basic assumption that the matrix and the activity are uniformly distributed in each segment, the classic model of SGS is built. Although the heterogeneity of the matrix and the activity could be reduced through rotating measurement of samples, it still does not satisfy with the basic assumption of model completely. The purpose of this work is to explore the effect of different density heterogeneity and position migration of emission source in SGS analysis for nuclear waste drum.

2 Principle

2.1 Transmission Measurement

According to the Beer-Lambert law [9], a parallel photon beam with energy E and intensity $I_0(E)$ penetrates through a material of thickness x in Eq. (1) as follows:

$$I(E) = I_0(E) \cdot \exp[-\mu(E) \cdot x] \quad (1)$$

where $I(E)$ is the intensity of transmitted photon and $\mu(E)$ is the linear attenuation coefficient. Because the count of every channel can be recorded in experimental gamma-ray spectrum, Eq. (1) transforms to Eq. (2):

$$N(E) = N_0(E) \cdot \exp[(-\mu(E) \cdot x)] \quad (2)$$

where $N(E)$ and $N_0(E)$ are the peak net count corresponding to energy E in the spectrum obtained with the empty and filled sample, respectively. The linear

attenuation coefficient $\mu(E)$ corresponded to each energy E is derived from $N(E)$ and $N_0(E)$ in Eq. (3) as follows:

$$\mu(E) = -\frac{1}{x} \cdot \ln \left[\frac{N(E)}{N_0(E)} \right] \quad (3)$$

Because $\mu(E)$ is related to energy of gamma ray, it is recommended to produce a fitted curve between the energy and the linear attenuation coefficient. Using this curve, the linear attenuation coefficient of gamma ray could be easily obtained for most of samples to be tested.

2.2 Emission Measurement

The emission measurement problems of SGS could be described with Eqs. (4)–(6) as follows:

$$D(E) = F(E) \cdot S(E) \quad (4)$$

$$F(E) = \varepsilon(E) \cdot A(E) \quad (5)$$

$$A(E) = \exp[-\mu(E) \cdot R] \quad (6)$$

where $D(E)$ is the detector's counting rate of sample, $F(E)$ is attenuation correction efficiency, $S(E)$ is the radioactivity of sample, $\varepsilon(E)$ is the detection efficiency, $A(E)$ is the self-absorption correction factor, R is the radius of sample. According to the fitted curve between the energy and the linear attenuation coefficient from transmission measurement, $\mu(E)$ of emission gamma ray could be obtained. Radioactivity of sample could be calculated as Eq. (7):

$$S(E) = \frac{D(E)}{\varepsilon(E) \cdot \exp[-\mu(E) \cdot R]} \quad (7)$$

3 SGS System

3.1 The Description of System

The fitted curve between the energy and the linear attenuation coefficient could be obtained through carrying out with rotating transmission scanning using SGS system. The self-absorption effect of sample could be corrected by the linear attenuation coefficient of emission gamma ray determined with this fitted curve. Radioactivity of sample could be determined through the experimental

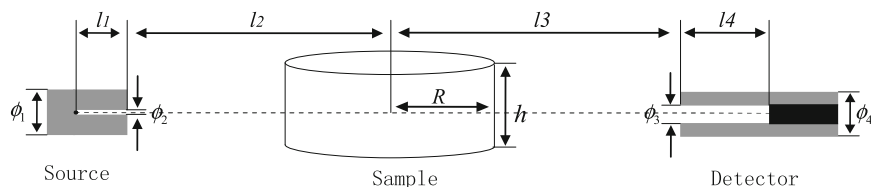
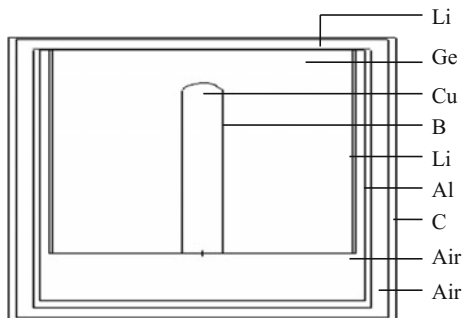


Fig. 1 Model of system

Fig. 2 Structure of HPGe detector



measurement and the self-absorption correction calculation. The model of SGS system is presented in Fig. 1.

The SGS system consists of the transmission source, sample and the detecting section as shown in Fig. 1. The length of transmission source collimator l_1 is 15 cm. Distance from the transmission source collimator's surface to the center of sample l_2 is 68.5 cm. Distance from the detector collimator's surface to the center of sample l_3 is 97 cm. The length of detector collimator l_4 is 18.5 cm. The height of sample h is 20 cm. The radius of sample R is 28 cm. The external diameter of transmission source collimator ϕ_1 is 19 cm. The internal diameter of transmission source collimator ϕ_2 is 1 cm. The internal diameter of detector collimator ϕ_3 is 8.7 cm. The external diameter of detector collimator ϕ_4 is 19 cm.

The electrical refrigeration P-type coaxial HPGe detector is used, and its structure is shown in Fig. 2. The diameter of the HPGe crystal is 7 cm, and its length is 8.26 cm. The diameter of cold finger copper is 0.9 cm and its length is 6.9 cm, the high voltage is positive 2600 V, operating temperature is 100 K, and the range of measured energy is from 4 keV to 10 MeV.

3.2 Samples

To explore the effect of different density heterogeneity for the nuclear waste drum samples in SGS analysis, several materials of different density are selected to correspond with actual contents of nuclear waste in the drum. Parameters of materials are presented in Table 1.

Table 1 Parameters of materials

Materials	Density (g.cm ⁻³)	Size (mm)
Hollow polyethylene	0.281	50 × 50 × 50
Solid polyethylene	1.037	50 × 50 × 50
Concrete	2.017	185 × 90 × 45
Glass fragments	1.438	Φ110 × 150
Fiber	0.206	Φ100 × 200
Plastics	1.411	160 × 160 × 85
Aluminum	2.762	150 × 100 × 60
Water	1.000	57 × 57 × 170

Table 2 Contents of eight samples

Number of sample	Contents
1#	Hollow polyethylene
2#	Solid polyethylene
3#	Solid polyethylene, glass fragments, plastics, water
4#	Hollow polyethylene, solid polyethylene, water
5#	Hollow polyethylene, solid polyethylene, water, glass fragments, plastics
6#	Hollow polyethylene, solid polyethylene, water, glass fragments, plastics, Concrete
7#	Hollow polyethylene, solid polyethylene, water, glass fragments, plastics, Concrete, Aluminum
8#	Hollow polyethylene, solid polyethylene, water, glass fragments, plastics, Concrete, Aluminum, Fiber

Using the several materials in Table 1 and grouping them with different methods, the eight samples of different density heterogeneity are built in SGS experimental measurement. The number and the actual contents of samples are presented in Table 2. The sources consist of ¹⁵²Eu(2.857 × 10⁸ Bq), ¹³⁷Cs(3.258 × 10⁵ Bq) and ⁶⁰Co(1.789 × 10⁵ Bq).

4 Results and Discussion

4.1 The Linear Attenuation Coefficients

Through rotating transmission scanning of the eight samples, the linear attenuation coefficients of different energy gamma ray are calculated with Eq. 3. The characteristic curves between the energy and the linear attenuation coefficient are fitted by the exponential function as shown in Fig. 3.

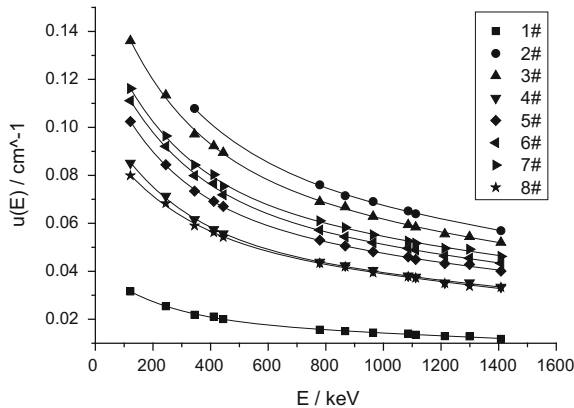


Fig. 3 Linear attenuation coefficient of gamma ray

Table 3 Linear attenuation coefficients of emission gamma ray

Energy (keV)	Linear attenuation coefficients (cm ⁻¹)							
	1#	2#	3#	4#	5#	6#	7#	8#
661.661	0.016898	0.081607	0.074695	0.047108	0.056604	0.061245	0.064947	0.046362
1173.238	0.013251	0.062565	0.057073	0.036684	0.044180	0.04758	0.050795	0.036133
1332.513	0.012402	0.058652	0.053414	0.034404	0.041540	0.044667	0.047768	0.033809

Because the sample 2# is constituted by the solid polyethylene and its thickness is large, it makes the corresponded gamma-photon can not be detected by HPGc detector. The linear attenuation coefficients of low-energy gamma ray of 121.7824 keV and 244.692 keV are lacked from characteristic curve of sample 2# shown in Fig. 3. For intermediate-energy gamma ray of 411.115, 443.976, 1212.95 and 1299.124 keV, because their respective emission branching ratios are 2.2, 3.04, 1.38 and 1.6%, they are so low that the counts of gamma-photon into detector are too little in the same transmission measurement time and a characteristic peak can not be presented. So the linear attenuation coefficients of those four energies gamma ray are lacked. Because the sample 1# is constituted by hollow polyethylene and its density is too low, it makes the lower linear attenuation coefficients than others.

For the emission gamma ray of 661.661, 1173.238 and 1332.513 keV, the linear attenuation coefficients could be determined by those fitted characteristic curves shown in Table 3.

4.2 Effect of Density

Radioactivity of samples could be determined through self-absorption correction calculation with Eq. 7. The correction radioactivity of the eight samples for

Table 4 Results of self- absorption correction of the eight radioactive samples

Number of samples	Self-absorption correction factors			Correction radioactivity (Bq)		
	661.661 keV	1173.238 keV	1332.513 keV	661.661 keV	1173.238 keV	1332.513 keV
1#	0.623040	0.690025	0.706624	3.176×10^5	1.799×10^5	1.804×10^5
2#	0.101774	0.173458	0.193543	3.163×10^5	1.802×10^5	1.802×10^5
3#	0.123507	0.202292	0.224116	3.194×10^5	1.812×10^5	1.810×10^5
4#	0.267396	0.358025	0.381627	3.289×10^5	1.827×10^5	1.837×10^5
5#	0.204966	0.290242	0.312510	3.392×10^5	1.844×10^5	1.849×10^5
6#	0.179989	0.263885	0.286311	3.372×10^5	1.851×10^5	1.862×10^5
7#	0.162266	0.241168	0.262500	3.387×10^5	1.852×10^5	1.869×10^5
8#	0.273040	0.363592	0.388038	3.392×10^5	1.841×10^5	1.868×10^5

661.661, 1773.238 and 1332.513 keV is presented in Table 4, and the relative deviations between corrected and actual radioactivity are shown in Fig. 4.

The relative deviations between corrected and actual radioactivity are less than 5% in eight samples of different density heterogeneity. With increasing of filling material density heterogeneity, the corrected radioactivity of samples and the relative deviations trend to increase gradually. It indicates that density heterogeneity determines the relative deviations directly in SGS analysis. Therefore, it is very important for reducing analysis deviation that known materials composition and distribution in the samples.

4.3 Effect of Position Migration

Figure 5 shows that the emission source is placed with equidistant migration ($d = 3.25$ cm) from the center to the border in the four samples 4# (0.541 g.cm^{-1}), 5# (0.636 g.cm^{-1}), 6# (0.684 g.cm^{-1}) and 7# (0.724 g.cm^{-1}). Eight distances from the center of sample to source, respectively, are 0, 3.25, 6.5, 9.75, 13, 16.25, 19.5, 22.75 cm. (a): Side view, (b): Vertical view.

The corrected activity of four samples for different emission source migration position is shown in Fig. 6. The density of sample 4# is low so that its corrected radioactivity is changed less in different migration position. For 661.661 keV, with average density and distance increasing gradually, the corrected radioactivity of samples 5#, 6# and 7# is obviously rising. The average density of samples is greater; the relative deviations between corrected and actual radioactivity seriously increase more. For 1173.238 and 1332.513 keV, because of their high energy and strong transmission ability, corrected activity has a rising trend but not obvious. Therefore, the density of samples, the energy and transmission ability of gamma ray are main effect factors of deviation in SGS analysis. It provides an effective idea to reduce the analysis deviation by reducing sample’s size or radius suitably and measuring low-density sample.

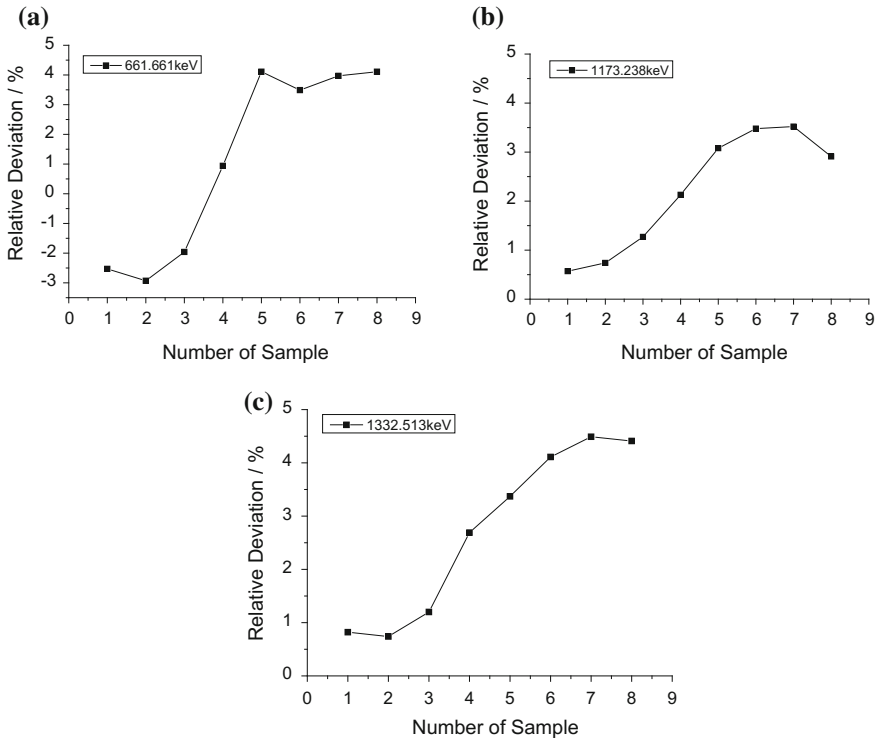
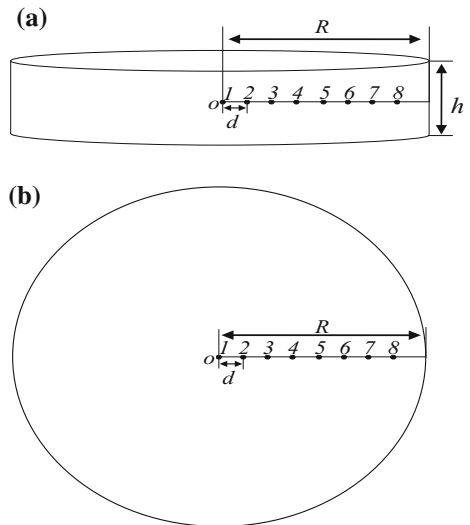


Fig. 4 Relative deviations between corrected and actual radioactivity of 661.661 keV (a) 1173.238 keV (b) and 1332.513 keV (c)

Fig. 5 Position migration of source



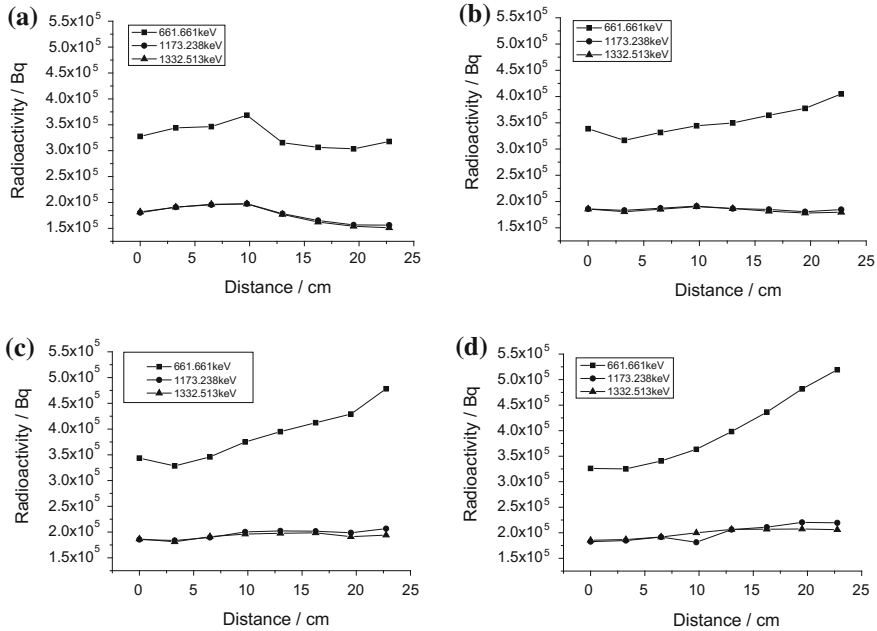


Fig. 6 Self-absorption correction results of samples 4# (a), 5# (b), 6# (c) and 7#(d)

5 Conclusions

The effect of deviation is studied in SGS analysis for different density heterogeneity nuclear waste sample segment. With filling material density heterogeneity and average density of samples increasing, the relative deviations between corrected and actual radioactivity trend to increase gradually. This method is suitable for low-density samples, and self-absorption correction analysis accuracy can be improved by reducing density of sample appropriately.

With source migrated from the center to the border in the samples, the relative deviations between corrected and actual radioactivity trend to increase gradually. The density of samples, the energy and transmission ability of gamma ray are main factors of deviation in SGS analysis. It provides an effective idea to reduce the analysis deviation by reducing sample's size or radius suitably and measuring low-density sample.

In order to improve the accuracy of the drum analysis, the tomographic gamma scanning (TGS) technique based on SGS has been put forward to analyze the nuclear waste drum [10, 11]. But considering with complexity of the TGS scanning measurement and the analysis method, we may need to explore another simple and effective method.

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