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## NEW MEASURING TECHNIQUE FOR ASSAY OF RADIOACTIVE MATERIALS IN WASTE DRUMS

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**Abstract:** A new measuring technique for determination of radioactive materials and estimate of the source distribution in waste drums is proposed. The principles of measurement and calculation results are presented.

Based on the mathematical simulation of the gamma ray measurement proposed in this paper with the Segmented Gamma Scanner calculations have been carried out. The calculation results prove that the new technique is much better than the well-known measurement method.

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### 1. INTRODUCTION

The characterization of radioactive waste is an important obligation in waste management and safeguards. Generally the methods for determination of the nuclide inventory rely on gamma-ray spectrometry or neutron counting. One of these methods is the Segmented Gamma Scanning technique (SGS), which is the important tool and widely has been used for assay of radioactive waste drums ( Bjork, 1987; Sprinkle and Hsue, 1987). SGS has been using the assumptions that the sample matrix and the radioactive source are uniform for a segment. However, these assumptions are generally not satisfied in practical cases. Nonuniform distribution of sample frequently causes the largest error ( Levai *et al*, 1995). Until now, this problem has not been solved although some techniques were proposed to detect and correct inhomogeneity in

the radioactive distribution (Cesana *et al*, 1993; Gillespie, 1994; Levai *et al*, 1995). In this paper a new measuring technique for determination of radioactive materials and estimate of the radioactive source distribution in waste drums is proposed. The principle and calculation results are presented. This technique has some advantages, as follow:

- In the case of uniform matrix, as will be shown below, the errors are small.
- Estimate of the radial distribution of radioactive sources for each segment is obtained.
- Answering the question why the traditional SGS technique has large systematic errors in case of nonuniform distribution of activity.
- Applicable to the traditional segmented gamma scanners with modifying only the software.

In addition, the errors caused by non uniformity of matrix are also estimated.

## 2. THEORETICAL BASIC

In a SGS system a radioactive waste drum is assayed in well defined vertical segments. If the measuring result for each segment is good the final result for the whole drum would be accurate. Therefore, it is necessary to consider relation between the count rate of detector and the distribution of radioactive materials in the matrix of a segment (see figure. 1)

Considering an isotope emitting gamma rays at energy  $E_i$ .

In the most general case, the count rate of detector caused by the isotope in a segment can be given as .

$$C = \alpha_i \int_0^R \int_0^{2\pi} I(r, \theta) \cdot f(r, \theta, \mu_i, K_j) r \cdot dr d\theta \quad (1)$$

where:

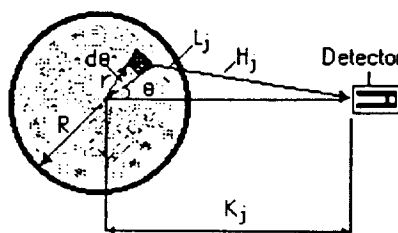


Figure 1: Measuring arrangement for a round source, a segment of drum. Count rate of detector caused by an element of the radioactive source in inhomogenous waste drum is considered.

$I(r, \theta)$  - a function representing the distribution of isotope of interest.

$f(r, \theta, \mu_i, K_j)$  - a function representing the geometry and absorption.

$$f(r, \theta, \mu_i, K_j) = \frac{\exp\left(-\int_0^{L_j} \mu_i(r, \theta) dL\right)}{H_j^2}$$

$\mu_i(r, \theta)$  - a function representing the distribution of the linear absorption coefficient corresponding to gamma ray energy  $E_i$ .

$L_j$  - path length of gamma ray in the absorber (waste mixture).

$$L_j = \frac{(R^2 \cdot H_j^2 - K_j^2 \cdot r^2 \cdot \sin^2 \theta)^{1/2} - (K_j \cdot \cos \theta - r) \cdot r}{H_j}$$

$K_j$  - distance from the detector to the center of segment, and

$$H_j = (K_j^2 + r^2 - 2K_j r \cos \theta)^{1/2}$$

$\alpha_i = \gamma_i \cdot \beta_i$ ,  $\gamma_i$  - yield of gamma line  $E_i$  of isotope;  $\beta_i$  - a function of the gamma ray energy and characteristic of the detector, which can be determined as  $\beta_i = \varepsilon_i \cdot K_{d,i}$  being  $\varepsilon_i$  the efficiency of a point source at the fixed distance  $K_{d,i}$ .  $R$  - Radius of drum.

In order to determine the activity of isotopes of interest and the radial distribution of the source, an approximate method is proposed. This method is based on the following principles and assumptions.

### ***Principles:***

- Rotate the drum during measurement.
- Divide the segment into a series of smaller rings.
- Measure the segment with different geometry by changing the distance from the detector to the center of segment ( $K_j$ ) and/or some different  $\gamma$  energy lines of the isotope of interest.
- Calculate the content of isotopes of interest of each ring source, and the results are summed to give the result for the segment.

### ***Assumptions:***

1. The linear absorption coefficient distribution  $\mu_i(r, \theta)$  is uniform, i.e. the absorber in the segment is homogeneous.
2. The distribution of radioactive source in each ring is uniform.
3. The function  $f(r, \theta, \mu_i, K_j)$  does not depend on the radius  $r$  in each ring and

$$f(r, \theta, \mu_i, K_j) = \exp(\mu_i L_j) / H_j^2 \quad (2)$$

The second and third assumptions are sensible because when the segment is rotated the dependence of function  $I(r, \theta)$  on angle  $\theta$  will lose, and the segment is divided into many rings so that the dependence of  $I(r, \theta)$  and  $f(r, \theta, \mu_i, K_j)$  on  $r$  can be ignored.

The first assumption generally is not satisfied in practice. The errors caused by this assumption will be estimated.

Let's suppose that the isotope emits  $m$  gamma energy lines.  $E_1, E_2 \dots E_m$  that correspond to linear absorption coefficients  $\mu_1, \mu_2, \dots, \mu_m$  of the absorbers. The segment is divided into  $n$  rings having the inner and outer radius  $(R_1, R_2), (R_2, R_3), \dots, (R_{n-1}, R_n)$ . The contents of the isotope in each ring are called  $I_1, I_2, \dots, I_n$ . From the above principles and assumptions, the count-rate  $C_1$  of the detector at distance  $K_1$  and gamma energy line  $E_1$  can be given, as follows.

By using the first and second assumptions, the equation (1) becomes.

$$C_1 = \frac{\alpha_1 I_1}{\pi(R_1^2 - R_0^2)} \int_{R_0}^{R_1} dr \int_0^{2\pi} f(r, \theta, \mu_1, K_1) r d\theta + \frac{\alpha_2 I_2}{\pi(R_2^2 - R_1^2)} \int_{R_1}^{R_2} dr \int_0^{2\pi} f(r, \theta, \mu_1, K_1) r d\theta +$$

$$+ \dots + \frac{\alpha_n I_n}{\pi(R_n^2 - R_{n-1}^2)} \int_{R_{n-1}}^{R_n} dr \int_0^{2\pi} f(r, \theta, \mu_1, K_1) r d\theta$$

By using the third assumption, eq. (3) becomes:

$$C_1 = \frac{\alpha_1 I_1}{2\pi} \int_0^{2\pi} f(R_0, \theta, \mu_1, K_1) d\theta + \frac{\alpha_1 I_2}{2\pi} \int_0^{2\pi} f(R_1, \theta, \mu_1, K_1) d\theta + \dots$$

$$+ \dots + \frac{\alpha_n I_n}{2\pi} \int_0^{2\pi} f(R_{n-1}, \theta, \mu_1, K_1) d\theta \quad (4)$$

The count-rates  $C_2, C_3, \dots, C_m$  of detector corresponding to gamma energy lines  $E_2, E_3, \dots, E_m$  can be expressed similarly to eq. (4), and



### 3. CALCULATION RESULT:

The aim of calculation work is to evaluate the performance of this method.

Based on the mathematical simulation of gamma ray measurements with the Segmented Gamma Scanner calculations have been carried out. Effects of the following parameters on the error have been estimated,

- the nonuniformity of the source.
- the density of matrices.
- the nonuniformity of matrices .
- number of the dividing rings.

A standard 200 waste drum with a diameter of 58 cm is modelled. Cases for homogeneous and inhomogeneous matrices and sources have been calculated. The average linear attenuation coefficients of 0.03, 0.06 and 0.12 cm<sup>-1</sup> are assumed. In order to simplify the problem without losing the generality here  $\alpha_i$  and the total content ( I ) of source are assumed equal to 1. All results are given as the relative ratios of the reading values to the true values. For the case in which a segment is measured at one distance and one gamma ray line (i.e. the segment is not divided, n=1) the well-known formula for correction of the attenuation

$$\frac{1 - \exp(-0.823\mu \cdot 2R)}{0.823\mu \cdot 2R}$$

have been used. Parameters for calculation are given in Table I:

*Case 1: radioactive source: point source; matrix: uniform*

Calculations have been made for measuring the extreme cases: a point source is located at specified radial positions in the drums. The results can be seen in Table II.

When the segment is divided into 4 rings, the errors are very small. For low and medium density matrix, the maximum error can be 1.1 %. For high density matrix ( $\mu=0.12 \text{ cm}^{-1}$ ), the maximum error can be 7.1 %. For low and medium density matrix, the segment divided only into 2 rings gives the acceptable errors ( $< 25 \%$ ). The effect of the number of the ring ( $n$ ) on the error is clear in the range of 1-4.

Table I: The parameters for calculation. Note: For the case  $n=1$ , i.e. the segment is not divided, the value  $R_k$  is not necessary to determined; the radius of drum  $R=29 \text{ cm}$ .

$n$	$\mu_i (\text{cm}^{-1})$	$K_j (\text{cm})$	$R_k (\text{cm})$
1	0.03	90	-
	0.06	90	-
	0.12	90	-
2	0.03	50; 90	0; 29
	0.06	50; 90	0; 29
	0.12	50; 90	0; 29
3	0.03	50; 70; 90	0; 26; 29
	0.06	50; 70; 90	0; 26; 29
	0.12	50; 70; 90	0; 26; 29
4	0.03	50; 60; 70; 90	0; 21; 27; 29
	0.06	50; 60; 70; 90	0; 21; 27; 29
	0.12	50; 60; 70; 90	0; 21; 27; 29
4	0.03; 0.04	50; 90	0; 21; 27; 29
	0.06; 0.07	50; 90	0; 21; 27; 29
	0.12; 0.13	50; 90	0; 21; 27; 29
6	0.03; 0.04	50; 70; 90	0; 15; 22; 26; 28; 29
	0.06; 0.07	50; 70; 90	0; 15; 22; 26; 28; 29
	0.12; 0.13	50; 70; 90	0; 15; 22; 26; 28; 29
9	0.12; 0.13; 0.14	50; 70; 90	0; 10; 16; 21; 24; 26; 27; 28; 29



Table II: Results for a point source at different positions (r), the absorber is uniform

$\mu$ (1/cm)	n	r (cm)							
		0	5	10	15	20	23	26	29
0.12	6	1.00	1.01	1.00	0.99	1.01	1.00	1.00	1.00
	4	1.00	1.01	1.02	0.99	0.99	1.04	1.08	1.00
	3	1.00	1.06	1.25	1.49	1.63	1.48	1.00	1.00
	2	1.00	1.10	1.42	2.01	2.89	3.19	2.95	1.00
	1	0.18	0.20	0.29	0.48	0.91	1.39	2.22	4.17
0.06	6	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
	4	1.00	1.00	0.99	0.99	1.00	1.01	1.01	1.00
	3	1.00	1.01	1.04	1.08	1.09	1.06	1.00	1.00
	2	1.00	1.03	1.10	1.21	1.33	1.35	1.26	1.00
	1	0.53	0.56	0.64	0.80	1.06	1.31	1.68	2.35
0.03	6	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
	4	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
	3	1.00	1.00	1.01	1.02	1.02	1.01	1.01	1.00
	2	1.00	1.02	1.10	1.21	1.33	1.35	1.26	1.00
	1	0.79	0.80	0.85	0.94	1.01	1.20	1.35	1.61

*Case 2: Radioactive source: ring sources; matrix: uniform*

In this case, the ring sources with the different inner and outer radius have been considered. Results can be seen in Table III. It is necessary to emphasized here that even if the source and matrix are uniformly distributed (see the final column) dividing the source into some small rings gives better results.

Table III: Results for a ring source with the different inner ( $r_1$ ) and outer ( $r_2$ ) radius in the uniform absorber

$\mu$ (1/cm)	$n$	$r_1 - r_2$ (cm)			
		0.-14.5	14.5-29.	8.-21.	0.-29.
0.12	4	0.97	1.05	0.95	1.01
	3	1.25	1.30	1.48	1.28
	2	1.45	2.62	2.13	2.33
	1	0.30	1.63	0.57	1.30
0.06	4	0.99	0.96	0.99	0.97
	3	1.04	1.04	1.08	1.05
	2	1.09	1.26	1.23	1.22
	1	0.65	1.38	0.84	1.20
0.03	4	1.00	1.00	0.99	1.00
	3	1.01	1.01	1.02	1.04
	2	1.03	1.07	1.06	1.05
	1	0.86	1.21	0.96	1.12

*Case 3: radioactive source: point source; matrix: non uniform.*

This is the worst case for all of the waste assay system. For simulating the non uniform distribution of matrices, the segment is divided into 4 parts, each with different attenuation. The average attenuation coefficients have been used to calculate the factor  $G_{kh}$  by expression (7). The average attenuation coefficients have been calculated by simulating the transmission technique that has been widely applied in SGS systems. The high and medium inhomogeneity of absorber has been considered

here. For the non uniform radioactive distribution, a point source is placed in a non-radioactive matrix.

Results in Table IV demonstrate that:

- In the case of medium inhomogeneity: for high density matrix the errors can be in range of 17 % - 36 %, for low and medium density they can be 2 % - 17 %.

-In the case of high inhomogeneity: the errors can be in the range of 52 % - 68 %, 25 % - 35 % and 7 % - 17 % for the high, medium and low density, respectively.

Table IV: Results for a point source at different positions ( $r$ ), the inhomogeneity of absorber is high and medium.

a) Medium inhomogeneity of gamma ray absorber:

$$\mu_1 = 2 \cdot \mu_4, \mu_2 = \mu_3 = 1.5 \cdot \mu_4$$

$\mu$ (1/cm)	n	r (cm)							
		0	5	10	15	20	23	26	29
0.12	4	1.38	1.57	1.60	1.51	1.55	1.80	1.87	1.07
	3	1.37	1.71	2.04	2.31	2.35	2.08	1.48	1.28
	2	1.38	1.80	2.34	3.05	3.84	4.01	3.67	1.57
	1	0.24	0.33	0.48	0.72	1.19	1.70	2.50	4.26
0.06	4	1.09	1.17	1.19	1.19	1.19	1.20	1.18	1.10
	3	1.09	1.20	1.27	1.31	1.30	1.24	1.15	1.09
	2	1.09	1.22	1.35	1.47	1.57	1.56	1.43	1.12
	1	0.58	0.67	0.79	0.96	1.23	1.47	1.81	2.41
0.03	4	1.02	1.06	1.07	1.08	1.08	1.08	1.07	1.05
	3	1.02	1.10	1.10	1.11	1.11	1.09	1.06	1.05
	2	1.02	1.08	1.12	1.16	1.18	1.17	1.13	1.05
	1	0.80	0.86	0.93	1.02	1.15	1.27	1.41	1.67

Table IV (continued)      b) High inhomogeneity of gamma ray absorber :

$$\mu_1 = 4\mu_4, \mu_2 = 2\mu_4, \mu_3 = 3\mu_4$$

$\mu$ (1/cm)	n	r (cm)							
		0	5	10	15	20	23	26	29
0.12	4	2.67	3.09	3.11	2.86	2.98	3.51	3.58	2.10
	3	2.67	3.32	3.81	4.03	3.77	3.26	2.41	1.92
	2	2.67	3.53	4.38	5.20	5.79	5.72	4.79	2.13
	1	0.47	0.66	0.89	1.22	1.74	2.23	2.96	4.38
0.06	4	1.32	1.49	1.55	1.54	1.52	1.54	1.49	1.33
	3	1.33	1.53	1.65	1.69	1.63	1.53	1.38	1.26
	2	1.33	1.58	1.76	1.90	1.94	1.87	1.68	1.27
	1	0.70	0.87	1.04	1.23	1.50	1.73	2.02	2.50
0.03	4	1.07	1.16	1.20	1.20	1.20	1.19	1.18	1.13
	3	1.08	1.17	1.22	1.24	1.23	1.20	1.16	1.11
	2	1.08	1.18	1.25	1.30	1.31	1.28	1.21	1.11
	1	0.85	0.95	1.04	1.15	1.28	1.39	1.51	1.70

Besides determination of the total content, this measuring technique allows the estimate of source distribution in a section of drum. Good results can be obtained for uniform matrices. Table V illustrates some cases. By comparison between  $n=4$  and  $n=9$ , it is clear that the more part a segment is divided into, the more exactly the positions of the sources are estimated.

#### 4. DISCUSSION

The problem for determination of radioactive material content in entirely unknown drum is difficult because the errors depend on many parameters in which the distribution of radioactive materials and matrix is the most important. Mathematically, the system of equations (6) is more general than the traditional formula being used in SGS technique so that it gives better results. This can be proved, as follows.

Table V: estimate of positions of point sources

a)  $n=4$  , the segment is divided into 4 parts.

Source	Distribution of gamma sources along radius (cm)				Total content	Conclusion
	0	21	27	29		
true	source at 0 cm				1	source at 0 cm
estimated	1.000	0.000	0.000	0.000	1.000	
true	source at 5 cm				1	source in the range 0-21 cm
estimated	0.955	0.079	-0.039	0.013	1.007	
true	source at 10 cm				1	source in the range 0-21 cm
estimated	0.789	0.325	-0.149	0.050	1.015	
true	source at 15 cm				1	source in the range 0-21 cm
estimated	0.456	0.721	-0.264	0.085	0.999	
true	source at 20 cm				1	source at 21 cm
estimated	0.052	1.014	-0.111	0.033	0.989	
true	source at 29 cm				1	source at 29 cm
estimated	0.000	0.000	0.000	1.000	1.000	

Considering the specified case of  $n=1$ , i.e. the segment is not divided and measured at one distance  $K_1$  ( $j=1$ ) and one gamma ray energy line ( $m=1$ ). Using the assumption of uniform radioactive material and matrix, and  $K$  longer than  $R$  many times, then the system (6) becomes the single equation as

$$C \approx \frac{\alpha I}{K^2} \frac{1}{\pi R^2} \int_0^R dr \int_0^{2\pi} \exp[-\mu(R^2 - r^2 \sin^2 \theta)] r d\theta \approx$$

$$\approx \frac{\alpha I}{K^2} \frac{1 - \exp(-0.823\mu \cdot 2R)}{0.823 \cdot \mu \cdot 2R} \quad (8)$$

Table V (continued)      b)  $n=9$ , the segment is divided into 9 parts.

Source	Distribution of gamma sources along radius (cm)									Total content	Conclusion
	0	10	16	21	24	26	27	28	29		
true	source at 0 cm									1	source at 0 cm
estimated	1.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	1.00	
true	source at 5 cm									1	source in the range 0-10 cm
estimated	0.649	0.440	-0.108	0.013	0.031	-0.058	0.042	-0.008	0.000	1.00	
true	source at 10 cm									1	source at 10 cm
estimated	0.000	1.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	1.00	
true	source at 15 cm									1	source in the range 10-16 cm
estimated	-0.061	0.247	0.875	-0.073	0.008	-0.024	0.022	-0.005	0.000	1.00	
true	source at 20 cm									1	source in the range 16-21 cm
estimated	0.061	-0.176	0.343	0.871	-0.116	0.009	0.015	-0.005	0.000	1.00	
true	source at 29 cm									1	source at 29 cm
estimated	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	1.000	1.00	

The equation (8) is the well-known formula that has been widely used to estimate activity in SGS. This shows that the results given by application of formula (8) have big errors because a too rough approximation ( $n=1$ ) has been used. In short, the traditional measurement is the most simple case of this method.

The division of a segment leads to estimate the distribution of source. As seen in Table 5, the results give the negative values at some positions. This can be explained from mathematical point of view. The method proposed here is an approximate technique where the function demonstrating the distribution of source is approximated by the other function. So, they can not be the same at all points. Comparison between

results of  $n=4$  and of  $n=9$  shows that the results of  $n=9$  are much better than that of  $n=4$ .

In principle, the more part a segment is divided into, the more correct the results are. However, as seen above, when  $n$  is equal to 6 the errors become very small. So, the increase in the number of parts has a meaning for estimate of the source distribution only.

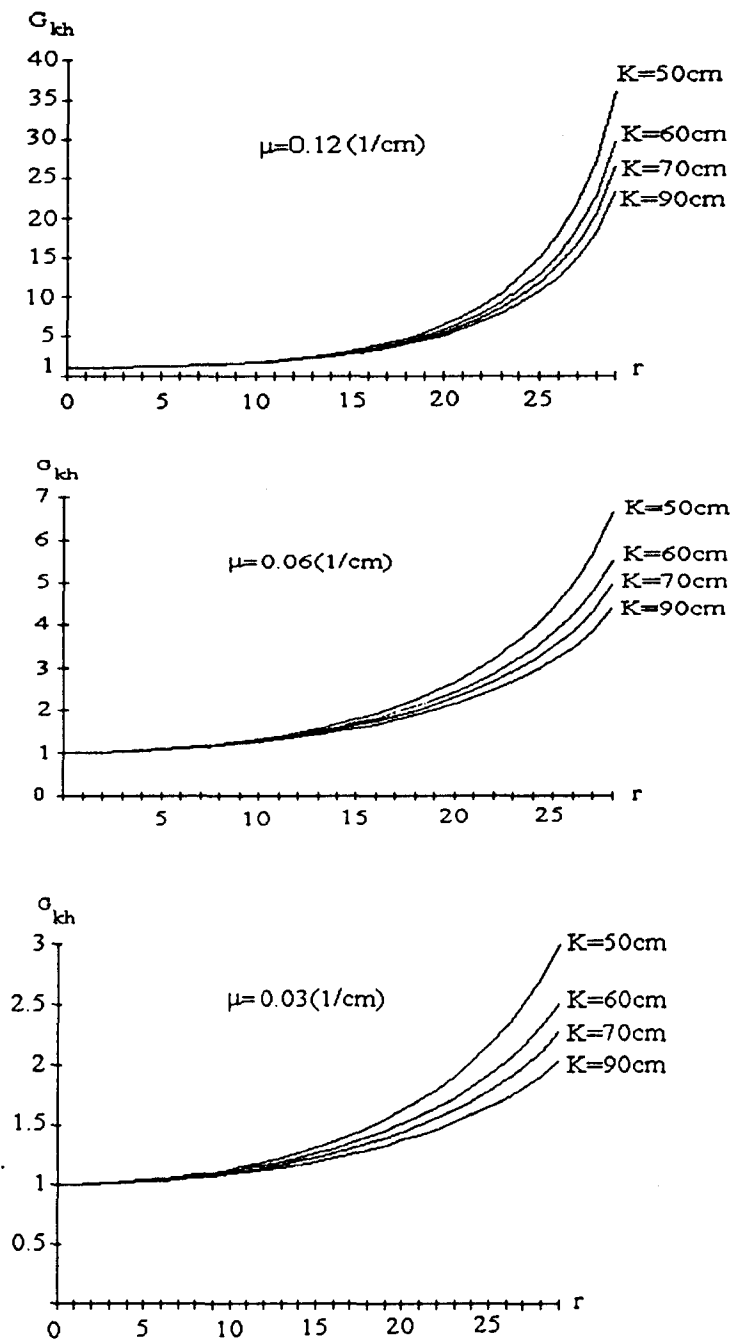
Procedure for dividing the segments, i.e. determining the elements  $G_{kh}$  of the matrix in the system (6), has an effect on the accuracy of results. Figure 2 illustrates the elements  $G_{kh}$  as functions of the absorption coefficient, the distance  $K_j$  and the radius of the dividing rings  $R_k$ . For the fixed  $\mu_i$  and  $K_j$ , they increase slowly when radii are smaller than 12 cm, but very fast when  $R_k$  is bigger than 20 cm. Furthermore, the higher the  $\mu_i$  is the faster they increase. For distances  $K_j$  bigger than 90 cm, they vary slowly. The longer the distance  $K_j$  is, the smaller the change is. From these comments on characteristics of the factor  $G_{kh}$ , some guidelines to the procedure for dividing segment and measurement can be given, as follows.

- Segments should not be measured at distances  $K_j$  bigger than 120 cm
- The optimum number of rings depends on the attenuation coefficients. A higher attenuation coefficient requires the more rings.
- The nearer a part of segment closes the perimeter the more rings it should be divided into.

Contrary to the traditional measurements where increasing the sample - to - detector distance is used to reduce errors caused by nonuniformity of sample this technique works in a close geometry experimentally taking into account absorption and geometry coefficients. This is useful to reduce measuring time and statistical errors for low activity samples.

Figure.2: The dependence of the factor  $G_{kh}$  on the positions of source ( $r$ ), the attenuation coefficients ( $\mu$ ) and the sample -to -detector distances ( $K$ ).

Note: Normalized scales.





In order to solve the system (6), the count-rates at the different distances are the important parameters. During the measurements the errors of the count-rates is not avoided. Fortunately, the errors of the activity measurements caused by inaccurate count-rates are small. Investigations show that a systematic error of 0.5 % - 1.5 % would be added if the errors of count rate are in the range 5 % - 30 %. (with  $n = 4$  )

## 5. CONCLUSIONS

In principle, the measuring procedure proposed in this paper solves successfully the problem of nonuniform distribution of radioactive materials in waste drum.

The determination of the gamma ray activity in concrete barrels (i.e. homogeneous matrix drums) is a demanding task although a method has been described (Filss, 1989). That technique may give large errors as the assumption of homogeneous distribution of the radioactive materials is still used. This method allows to solve this problem with high accuracy.

As far as systematic errors are concerned, this method gives better results than that of the traditional SGS because the assumption of homogenous distribution of the radioactive materials is excluded. More over, it allows to estimate the radial distribution of activity in each segment.

Although the assumption of homogeneous matrix in this method is not satisfied in practice, the above investigations show that it can be used for the case of inhomogeneous matrix having low and medium density ( $\mu=0,01-0.06 \text{ cm}^{-1}$ ).

In practice, this technique can be applied immediately for the SGS system with minor software modifications.

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