Available online at www.sciencedirect.com
SCIENCE DIRECT®
ELSEVIER
Annals of Nuclear Energy 32 (2005) 1516–1523
annals of NUCLEAR ENERGY
www.elsevier.com/locate/anucene

Technical note

Evaluation of performance of a new gamma technique for assay of radioactive waste

Tran Ha Anh, Nguyen Duc Thanh, Tran Quoc Dung *

Centre for Nuclear Techniques, 217 Nguyen Trai, D1, Hochiminh City, Vietnam

Received 28 January 2005; accepted 29 March 2005
Available online 31 May 2005

Abstract

In order to increase the accuracy of measurement of radioactive waste drum assay by Segmented Gamma Scanning technique a new measurement technique was proposed. Results of the study with noise data of measuring factors are presented. The results confirm that the technique can be successfully developed for assay of radioactive waste drums.

1. Introduction

The Segmented Gamma Scanner (SGS) is an important tool for isotopic composition measurement and for determination of the activity in gamma contaminated waste drums (Bjork, 1987; Sprinkle and Hsue, 1987). SGS has been used with the assumptions that: (a) both the sample matrix and the radioactive source are uniform; (b) the γ-ray transmission through drums is not smaller than 0.2. However, these are generally not satisfied in practical cases. The systematic error of this technique is still large due to the following reasons: (a) non-uniform distribution of radioactive source within the drums frequently causes the largest error (Estep, 1990; Dung, 1997); (b) non-uniform distribution of non-radioactive materials (matrix) (Gillespie, 1994; Dung, 1997); (c) particle size of the nuclear material, the lump effect, specially for

* Corresponding author. Tel.: +848 8368865; fax: +848 8367361.
E-mail address: dungtranquoc@gmail.com (T.Q. Dung).

0306-4549/$ - see front matter
doi:10.1016/j.anucene.2005.03.010

uranium and plutonium assay (Sprinkle and Hsue, 1987; Prettyman et al., 1996; Anh and Dung, 2001); the drum-to-detector distance (Dung, 1997).

In order to increase measurement accuracy a new measurement technique for determination of radioactive materials and estimation of radioactive source distribution in waste drums was studied (Dung, 1998a,b). Some new results were presented (Dung, 2004). This technique has some advantages, such as: the errors are small in the case of uniform matrix; the estimation of the radial distribution of radioactive sources for each segment is obtained; the question of why the traditional SGS technique has large systematic errors in case of non-uniform distribution of activity is answered; the traditional segmented gamma scanners is applicable with modifying only the software. In order to evaluate the performance of this method for practical application noise data of measuring factors are considered by simulating the measurement system. The results given in this paper confirm that this technique can be successfully developed for assay of radioactive waste drums in practice.

2. Theoretical basis

In a SGS system a radioactive waste drum is assayed in well-defined vertical segments. If the measurement result for each segment were good the final result for the whole drum would be accurate. The idea of this method is that the segment is divided into several rings (see Fig. 1), and measured with some different gamma energy lines of the isotope of interest and/or at different distances from the detector to the center of segment. So the parameters relating to the detector-to-drum distance and gamma energies are subscripted by j and i, respectively. This method is based on the following principles and assumptions (Dung, 1998a,b):

Principles: (1) rotate the drum during measurement; (2) divide the segment into a series of smaller rings; (3) measure the segment with different geometry by changing the distance from the detector to the center of segment and/or some different γ energy lines of the isotope of interest; (4) calculate the content of isotope 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 is uniform, i.e., the absorber in the segment is homogeneous; (2) the distribution of radioactive source in

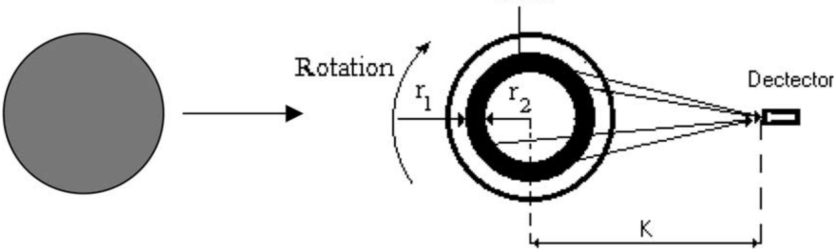

Fig. 1. The segment is divided into several rings and measured with some different gamma energy lines of the isotope of interest and/or at different distances from the detector to the center of segment.

each ring is uniform; (3) the function representing the geometry and absorption does not depend on the radius r in each ring.

If the segment is divided into n rings having the inner and outer radius (R_0, R_1), (R_1, R_2), (R_2, R_3), ..., (R_{n-1}, R_n). The contents of the isotope in each ring are called I_1, I_2, ..., I_n. From these principles and assumptions, the relation between the content of the isotope of I_1, I_2, ..., I_n in each ring and the count-rates of detector can be described by the following equation system:

$$\begin{array}{ll}
m \text{ equations} & C_1 = I_1G_{11} + I_2G_{12} + \cdots + I_nG_{1n} \\
\multicolumn{2}{l}{\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots} \\
\text{for } K_1 & C_m = I_1G_{m1} + I_2G_{m2} + \cdots + I_nG_{mn} \\
m \text{ equations} & C_{m+1} = I_1G_{(m+1)1} + I_2G_{(m+1)2} + \cdots + I_nG_{(m+1)n} \\
\multicolumn{2}{l}{\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots} \\
\text{for } K_2 & C_{2m} = I_1G_{(2m)1} + I_2G_{(2m)2} + \cdots + I_nG_{(2m)n} \\
\multicolumn{2}{l}{\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots} \\
\multicolumn{2}{l}{\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots} \\
m \text{ equations} & C_{n-m-1} = I_1G_{(n-m-1)1} + I_2G_{(n-m-1)2} + \cdots + I_nG_{(n-m-1)n} \\
\multicolumn{2}{l}{\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots} \\
\text{for } K_p & C_n = I_1G_{n1} + I_2G_{n2} + \cdots + I_nG_{nn}.
\end{array} \tag{1}$$

Here, the isotope in the segment is measured with m γ-ray lines and at p distances (K_j). G_{kh} – coefficients that depend on the geometry and absorption in each ring. C_l – count rates of detector

$$G_{kh} = \frac{\alpha_i}{2\pi}\int_0^{2\pi} f(R_{k-1}, \theta, \mu_i, K_j)\,\mathrm{d}\theta, \tag{2}$$

where ($k = 1, 2, \ldots, n$; $h = i + (j-1).m$; $i = 1, 2, \ldots, m$, $j = 1, 2, \ldots, p$), α_i – a factor, which depends on the yield of gamma line E_i of isotope, the γ-ray energy and the characteristics of the detector, and

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

After determining C_k by measurements and G_{kh} by calculation, the content I_k will be given by solving the system (1). The values I_k in the kth ring show the radial distribution of the isotope in the segment. The sum of I_k gives the result for the segment considered. The total content of the isotope in the whole drum can be given as the sum of the results.

3. Results and discussion

Systematic errors can be obtained when these assumptions are not satisfied in practical application. In order to evaluate the performance of this method calculation work has been carried out by simulating a measurement system. A segment of standard 208-l waste drum with diameter of 58 cm is modeled. γ-Ray measurement is made at fission product isotope energies from 140 to 1400 keV, and average densities in range of 0.2–1 g/cm^3 resulting in average linear attenuation coefficients from 0.01 to 0.14 cm^{-1}. The average linear attenuation coefficients 0.03, 0.06 and 0.12 cm^{-1} are chosen.

In order to evaluate the performance of this method and compare with the traditional SGS technique, calculation work has been carried out by simulating a measurement system. The results are given in Table 1.

For practical applications noise data is also investigated. A point source that usually causes the largest error in SGS is considered (see Fig. 2). The least squares model

Table 1a
Results for a point source at different positions in homogeneous matrix

μ (cm^{-1})	Method	r (cm)							
		0	5	10	15	20	23	26	29
	SGS	0.18	0.20	0.29	0.48	0.91	1.39	2.22	4.17
0.12	This method	1.00	1.01	1.02	0.99	0.99	1.04	1.08	1.00
	SGS	0.53	0.56	0.64	0.80	1.06	1.31	1.68	2.35
0.06	This method	1.00	1.00	0.99	0.99	1.00	1.01	1.01	1.00
	SGS	0.79	0.80	0.85	0.94	1.01	1.20	1.35	1.61
0.03	This method	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00

Note. The segment was divided into four rings.

Table 1b
Results for a point source at different position (r), the in homogeneity of absorber is high and medium

μ (cm^{-1})	Method	r (cm)							
		0	5	10	15	20	23	26	29
Medium inhomogeneity of γ-ray absorber									
	SGS	0.24	0.33	0.48	0.72	1.19	1.70	2.50	4.26
0.12	This method	1.38	1.57	1.60	1.51	1.55	1.80	1.87	1.07
	SGS	0.58	0.67	0.79	0.96	1.23	1.47	1.81	2.41
0.06	This method	1.09	1.17	1.19	1.19	1.19	1.20	1.18	1.10
	SGS	0.80	0.86	0.93	1.02	1.15	1.27	1.41	1.67
0.03	This method	1.02	1.06	1.07	1.08	1.08	1.08	1.07	1.05
High inhomogeneity of γ-ray absorber									
	SGS	0.47	0.66	0.89	1.22	1.74	2.23	2.96	4.38
0.12	This method	2.67	3.09	3.11	2.86	2.98	3.51	3.58	2.10
	SGS	0.70	0.87	1.04	1.23	1.50	1.73	2.02	2.50
0.06	This method	1.32	1.49	1.55	1.54	1.52	1.54	1.49	1.33
	SGS	0.85	0.95	1.04	1.15	1.28	1.39	1.51	1.70
0.03	This method	1.07	1.16	1.20	1.20	1.20	1.19	1.18	1.13

Table 1c
Results for an extensive source in homogenous matrix within in a segment

Method	ϕ	R (cm)							
		3.625	7.25	10.875	14.5	18.125	21.75	25.375	29
(a) $\mu = 0.03$ cm^{-1}									
SGS		0.79	0.80	0.83	0.86	0.90	0.96	1.02	1.13
This method	360°	1.00	1.00	1.00	0.99	0.99	0.98	1.01	1.02
SGS		0.79	0.80	0.83	0.86	0.90	0.96	1.02	1.13
This method	270°	1.00	1.00	1.00	0.99	0.99	0.98	1.01	1.02
SGS		0.79	0.80	0.83	0.86	0.90	0.96	1.02	1.13
This method	180°	1.00	1.00	1.00	0.99	0.99	0.98	1.01	1.02
SGS		0.79	0.80	0.83	0.86	0.90	0.96	1.02	1.13
This method	90°	1.00	1.00	1.00	0.99	0.99	0.98	1.01	1.02
$\mu = 0.06$ cm^{-1}									
SGS		0.54	0.56	0.60	0.65	0.73	0.83	0.98	1.21
This method	360°	1.00	0.98	0.97	0.98	1.01	1.03	1.02	1.01
SGS		0.54	0.56	0.60	0.65	0.73	0.83	0.98	1.21
This method	270°	1.00	0.98	0.97	0.98	1.01	1.03	1.02	1.01
SGS		0.54	0.56	0.60	0.65	0.73	0.83	0.98	1.21
This method	180°	1.00	0.98	0.97	0.98	1.01	1.03	1.02	1.01
SGS		0.54	0.56	0.60	0.65	0.73	0.83	0.98	1.21
This method	90°	1.00	0.98	0.97	0.98	1.01	1.03	1.02	1.01
$\mu = 0.12$ cm^{-1}									
SGS		0.18	0.20	0.24	0.30	0.40	0.56	0.83	1.31
This method	360°	0.99	0.98	0.97	1.01	1.02	1.00	0.99	0.97
SGS		0.18	0.20	0.24	0.30	0.40	0.56	0.83	1.31
This method	270°	0.99	0.98	0.97	1.01	1.02	1.00	0.99	0.97
SGS		0.18	0.20	0.24	0.30	0.40	0.56	0.83	1.31
This method	180°	0.99	0.98	0.97	1.01	1.02	1.00	0.99	0.97
SGS		0.18	0.20	0.24	0.30	0.40	0.56	0.83	1.31
This method	90°	0.99	0.98	0.97	1.01	1.02	1.00	0.99	0.97

The total activity uniformly distributed in a sector of the segment is considered. ϕ, r – angle and radius of the sector.

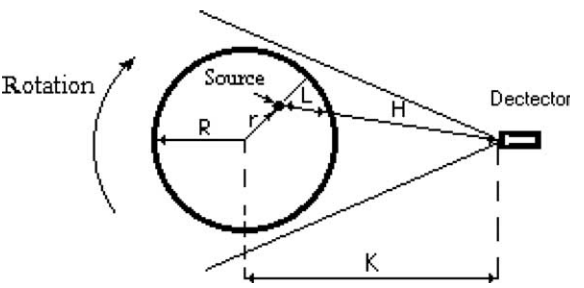

Fig. 2. A point source at different positions in the homogeneous matrix is considered.

Table 2
Results for a point source at different positions (r_n) with noise data

r_n	$e_{\mu 1}$	$e_{\mu 2}$	I_s	δ_s
(a) The error caused by noise data of the attenuation coefficients (μ)				
0.0	−5.	−5.	.98	−1.86
	−5.	5.	.90	−10.16
	10.	−10.	1.23	22.58
	20.	20.	1.07	7.40
	−20.	−20.	.93	−7.30
15.0	5.	5.	1.02	1.84
	5.	−5.	1.13	13.20
	−10.	10.	1.21	20.56
	20.	20.	1.06	5.84
	−20.	−20.	0.95	−5.42
29.0	5.	−5.	0.99	−1.13
	−5.	5.	0.96	−3.58
	10.	−10.	1.11	10.78
	20.	20.	1.04	4.05
	−20.	−20.	0.96	−4.14

r_n	e_{K1}	e_{K2}	I_s	δ_s
(b) The error caused by noise data of the drum to detector distances (K)				
0.0	−5.	−5.	0.90	−9.73
	−5.	5.	1.16	16.10
	10.	−10.	1.24	24.42
	20.	20.	1.44	43.95
	−20.	−20.	0.64	−36.00
15.0	5.	5.	1.11	10.67
	−5.	5.	1.16	16.05
	10.	−10.	0.44	−55.63
	20.	20.	1.43	43.23
	−20.	−20.	0.63	−37.38
29.0	−5.	−5.	0.84	−15.58
	−5.	5.	0.92	−8.07
	10.	−10.	1.19	19.46
	20.	20.	1.49	48.72
	−20.	−20.	0.35	−64.65

r_n	e_{C1}	e_{C1}	e_{C1}	e_{C1}	I_s	δ_s
(c) The error caused by noise data of the detector count rate						
0.0	5.	5.	5.	5.	1.05	0.51
	−5.	−9.	−5.	−9.	0.97	−2.7
	−10.	−10.	−10.	−10.	0.90	−9.97
	10.	18.	10.	18.	1.06	5.50
	−15.	−27.	−15.	−27.	0.92	−8.22
	20.	20.	20.	20.	1.20	20.01

(*continued on next page*)

Table 2 (*continued*)

r_n	e_{C1}	e_{C1}	e_{C1}	e_{C1}	I_s	δ_s
15.0	−5.	−5.	−5.	−5.	0.95	4.57
	5.	9.	5.	9.	1.02	2.48
	10.	10.	10.	10.	1.11	10.50
	−10.	−18.	−10.	−18.	0.96	−3.65
	15.	27.	15.	27.	1.07	6.62
	−20.	−20.	−20.	−20.	80.0	−19.66
29.0	5.	5.	5.	5.	1.05	4.93
	−5.	9.	−5.	9.	0.97	−2.88
	10.	−18.	10.	−18.	1.06	5.73
	10.	18.	10.	18.	1.00	0.06
	−15.	−27.	−15.	−27.	1.00	0.44
	−10.	−10.	−10.	−10.	1.09	8.56
	20.	20.	20.	20.	1.20	19.63

r_n	$e_{\mu1}$	$e_{\mu2}$	e_{K1}	e_{K2}	e_{C1}	e_{C1}	e_{C1}	e_{C1}	I_s	δ_s
(d) The error caused by noise data of the parameters										
0.0	2.	2.	2.	2.	2.	2.	2.	2.	1.07	6.89
	−5.	−5.	−5.	−5.	−5.	−5.	−5.	−5.	0.84	−15.67
	−5.	5.	−5.	5.	−5.	5.	−5.	5.	0.92	−8.25
	10.	−10.	2.	.	5.	5.	5.	5.	1.44	44.20
	10.	10.	10.	10.	10.	10.	10.	10.	1.39	39.07
	15.	15.	5.	5.	1.	1.	1.	1.	1.18	18.39
	−20.	−20.	2.	2.	5.	5.	5.	5.	1.01	0.79
	−20.	20.	2.	2.	5.	5.	5.	5.	0.77	−22.99
15.0	2.	2.	2.	2.	2.	2.	.	2.	1.07	7.24
	−5.	−5.	−5.	−5.	−5.	−5.	−5.	−5.	0.85	−15.09
	−5.	5.	−5.	5.	−5.	5.	−5.	5.	0.90	−10.32
	10.	−10.	2.	2.	5.	5.	5.	5.	1.42	41.79
	10.	10.	10.	10.	10.	10.	10.	10.	1.38	38.22
	10.	−10.	10.	−10.	10.	−10.	10.	−10.	1.29	30.56
	15.	15.	5.	5.	1.	1.	1.	1.	1.17	17.37
	−20.	20.	2.	2.	5.	5.	5.	5.	0.80	−19.83
29.0	2.	2.	2.	2.	2.	2.	2.	2.	1.09	8.56
	−5.	−5.	−5.	−5.	−5.	−5.	−5.	−5.	0.80	−20.15
	−5.	5.	−5.	5.	−5.	5.	−5.	5.	0.72	−28.15
	10.	−10.	2.	2.	5.	5.	5.	5.	1.23	22.54
	10.	10.	10.	10.	10.	10.	10.	10.	1.46	46.15
	10.	−10.	10.	−10.	10.	−10.	10.	−10.	0.79	20.50
	15.	15.	5.	5.	1.	1.	1.	1.	1.21	20.87
	−20.	20.	2.	2.	5.	5.	5.	5.	1.00	0.2

Note. The "true" activity is supposed equal to 1.

I_s, δ_s – value of activity and error by using the least squares model for noisy data.

$e_{\mu1}$, $e_{\mu2}$ – random errors (%) of the attenuation coefficients.

e_{K1}, e_{K2} – random errors (%) of the segment-to-detector distances.

was used. Several simulations have been made, and main results are given in Table 2. It is easy to see that the noise data causes smaller errors in comparison with systematic errors. The investigation shows that this method gives the best results for all cases. Although the assumption of homogeneous matrix is not satisfied in practice, the above investigation shows that it can be used for inhomogeneous matrix having low and medium density (μ from 0.01 to 0.06 cm^{-1}).

4. Conclusion

The results demonstrate that for the estimation of activity concentrations large systematic errors can be introduced by traditional SGS because the measured values are affected by many factors. Therefore these errors should be estimated when assaying radioactive waste drums.

The measurement technique given above solves successfully the problem of non-uniform distribution of radioactive materials in waste drum. As far as systematic errors are concerned, this method gives better results than the traditional SGS because the assumption of homogenous distribution of the radioactive materials is excluded.

The results prove that the random errors are much smaller than the systematic errors, and that this technique can be successfully applied in practice for assay of radioactive materials.

Acknowledgements

This work was supported from the Vietnam Atomic Energy Commission. The authors deeply thank Bui Xuan Huy, Luu Anh Tuyen and Lo Thai Son for their help with elaboration of the calculation program.

References

Anh, T.H., Dung, T.Q., 2001. Ann. Nucl. Energy 24, 265–273.

Bjork, C.W. In: Proceedings of the Third International Conference on Facility Operation Safeguards Interface. Nov. 29–Dec. 4, San-Diego, CA, 1987, p. 129.

Dung, T.Q., 1997. Ann. Nucl. Energy 24 (1), 33–47.

Dung, T.Q., 1998a. Progr. Nucl. Energy 33 (4), 403.

Dung, T.Q., 1998b. Nucl. Inst. Meth. Phys. Res A 416, 505.

Dung, T.Q. In: Proceedings of the 9th Asia Pacific Physics Conference, 25–31 November, Hanoi, 2004.

Estep, R.J., 1990. Trans. Am. Nucl. Soc 62, 178.

Gillespie, B.M. In: IAEA Symposium on International Safeguards, 8–14 May, Vienna, 1994.

Prettyman, T.H., Foster, L.A., Estep, R.J. LA-UR-96-2620, 1996.

Sprinkle, J.K., Hsue, S.T. In: Proceedings of the Third International Conference on Facility Operation Safeguards Interface, Nov. 29–Dec. 4, San-Diego, CA, 1987, p. 188.