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ARTICLE

Relaxation-depth sensitivity of In-situ γ Spectrometry to Determine the Depth-distribution of Artificial Radionuclides in Soil

FENG Tiancheng[1,2*], JIA Mingyan[2], CHENG Jianping[1] and FENG Yuanju[2]

[1] *Department of Engineering Physics, Tsinghua University, Zijing Building 0816B, Beijing 100084, China*
[2] *Northwest Institute of Nuclear Technology, Xi'an, China, P.O.Box 69-1 Xi'an 710024, China*

The main limitation of in-situ γ spectrometry lies in determining the depth-distribution of the artificial radionuclide in soil. Many researchers have developed methods and models for deducing the depth-distribution information from in-situ spectrum itself. Until now, such methods were studied and established as "Multiple photopeak method", "Peak-to-valley ratio method" and "Collimation or lead-plate method". This paper presented the comparative theoretical study on the relaxation-depth sensitivity. The experimental methods for Multiple photopeak, Peak-to-valley ratio and Collimation or lead-plate are at energies of 244keV and 1408keV γ-rays of ^{152}Eu, 662keV γ-ray of ^{137}Cs, 662keV γ-ray of ^{137}Cs, respectively, and the In-situ object counting system was employed in Collimation or lead-plate method. Results indicated that the sequence of relaxation-depth sensitivity from the maximum to the minimum was as following: Peak-to-valley method, Multiple photopeak method, and Collimation or lead-plate method, and the sensitivity of Collimation or lead-plate method was far less than Multiple photopeak method.

KEYWORDS*: *in-situ γ spectrometry, Multiple photopeak method, Peak-to-valley method, Collimation or lead-plate method, relaxation-depth sensitivity

I. Introduction

In-situ HPGe γ spectrometry is a rapid and powerful method for the survey of radioactivity in environment. When it is used to determine the environmental radiation, initial assumptions are usually made about the depth-distribution of the radionuclide of interest in soil, in order to derive their activity concentration from the spectrum. Such assumptions can be a principal source of the uncertainty in the final results [1,2)]. Many researchers have developed methods and models for deducing the depth-distribution information from in-situ spectrum itself. Until now, such methods were studied and established as "Peak-to-valley ratio method" [3-9)], "Collimation or lead-plate method" [10,13)] and "Multiple photopeak method" [14-18)].

For radionuclides emitting γ-rays of several energies some parameters of depth-distribution may be determined by analysis of a single measurement based on the different absorption of γ-rays in the soil. That is due to known energy dependence of attenuation coefficient in soil. Rays with different energy are absorbed differently. Ordinarily, the depth-distribution of artificial radionuclides deposited in ground soil can be expressed approximately with an exponential function. Combining information from different energies enables us to assess the depth-distribution, which is the "Multiple photopeak method"; The deeper radionuclides distributed in soil, the more γ-rays attenuated by soil, therefore Compton effect would be strengthened, and the ratio of peak-to-valley decreases. Combining information from different ratios the depth-distribution information can be extracted, which is the "Peak-to-valley ratio method"; And using different collimators or shielding lead plates, the directional distribution of γ-rays can be determined which in turn provides information on the effective burial depth, which is the basis of the "Collimation or lead-plate method".

J MacDonald's researches[19,20)] showed that the Multiple photopeak method offers the best potential sensitivity, and the lead plate method shows the least sensitivity. But until now, only J MacDonald's researches have been published to show the relaxation-depth sensitivities of these methods. And our work showed that there are some differences in the relaxation-depth sensitivities comparing to MacDonald's conclusions. Therefore, in this paper, the theoretical study was performed particularly on the relaxation-depth sensitivities for these main three methods in a comparative form.

II. Theories

1. Multiple photopeak method

For an infinite radius plane-source at depth z_i in soil, based on Beck Eq. [1)] the full peak count rate in the in-situ spectrum can be calculated as following Eq. (1), and the detection efficiency ε is defined as Eq. (2).

$$n = \frac{A_z P_\gamma S_0}{2} \int_0^1 \frac{F(\cos\theta)}{\cos\theta} e^{-(\mu_a h + \mu_s z_i)/\cos\theta} d\cos\theta \tag{1}$$

$$\varepsilon = \frac{n}{A_z} = \frac{P_\gamma S_0}{2} \int_0^1 \frac{F(\cos\theta)}{\cos\theta} e^{-(\mu_a h + \mu_s z_i)/\cos\theta} d\cos\theta \tag{2}$$

Where n is the full peak count rate of the γ-ray in spectrum, s^{-1}. A_z is the unit volume activity of the plane-source with a thickness Δz (→0) at depth z_i in soil, Bq/cm^3. P_γ is the probability of the γ-ray emission, s^{-1}. S_0 is the effective-front-area of the detector to the γ-ray, cm^2. $F(\cos\theta)$ is the angular-response-function of the detector to the γ-ray, which is a function of the polar angle θ between the detector-symmetry-axis and a radioactive-unit-element in plane-source. μ_{jS} , μ_{jA} are the linear attenuation coefficients of the γ-ray in soil and air respectively, cm^{-1}. h is the height of detector above the ground, cm.

By the Eq. (2), ε at a certain depth z_i in soil can be calculated. Calculation results show that the relationship between ε and z_i corresponds with the exponential function well. The detection efficiencies of the 1st and 2th energy

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*Corresponding Auhor, Tel No: 86-10-51537354. E-mail: ftc07@mals.tsingha.edu.cn[1] γ-rays, ε(1) and ε(2), can be fitted by the Eqs. (3) and (4), respectively.

$$\varepsilon(1) = ae^{-bz} \tag{3}$$

$$\varepsilon(2) = ce^{-dz} \tag{4}$$

Where *a*, *c*, *b* and *d* all are constants. The *z* is the depth of the plane-source in soil, cm.

The full peak count rates of the 1st and 2th energy γ rays, *n*(1) and *n*(2), can be expressed by Eqs. (5) and (6), respectively.

$$n(1) = A_0 S \int_0^{\infty} e^{-\alpha z} a e^{-bz} dz = \frac{A_0 S a}{b+\alpha} \tag{5}$$

$$n(2) = \frac{A_0 S c}{d+\alpha} \tag{6}$$

Where A_0 is the activity concentration at the soil surface, Bq/cm^3; *α* is the reciprocal of the relaxation depth, cm^{-1}; And the detection area *S* is infinite, cm^2.

The ratio of the full peak count rates of the 1st energy to 2th energy, *R*(*α*), is calculated by the following Eq. (7), which depicts the relationship between the *R*(*α*) and *α* concisely.

$$R(\alpha) = \frac{n(1)}{n(2)} = \frac{a}{c} \cdot \frac{d+\alpha}{b+\alpha} \tag{7}$$

2. Collimation or lead-plate method

In-situ object counting system (ISOCS) is employed for sensitivity analysis of "Collimation or lead-plate method", by which the detection efficiency can be calculated. For the maximization of the efficiency difference among the different shielding conditions, collimators of "30d-50mm" and "90d-50mm~180d-50mm" were adopted.

Calculation results show that for an infinite radius plane-source at the discrete depth z_i in soil, the relationship between the ε' with collimator and z_i corresponds with the exponential function well, but there is a constant appended comparing the Multiple photopeak method above. The detection efficiencies of the 1st and 2th collimators, $\varepsilon'(1)$ and $\varepsilon'(2)$, can be fitted by the Eqs. (8) and (9), respectively.

$$\varepsilon'(1) = a'e^{-b'z} + k_1 \tag{8}$$

$$\varepsilon'(2) = c'e^{-d'z} + k_2 \tag{9}$$

Where *a*′, *c*′, *b*′, *d*′, k_1 and k_2 all are constants.

As above, given the depth-distribution expressed with the exponential function and the detection area *S*′, the full peak count rates of the 1st and 2th collimators, *n*′(1) and *n*′(2), can be expressed by Eqs. (10) and (11), respectively.

$$n'(1) = A_0 S' \int_0^{\infty} e^{-\alpha z}(a'e^{-b'z} + k_1)dz = A_0 S'\left(\frac{a'}{b'+\alpha} + \frac{k_1}{\alpha}\right) \tag{10}$$

$$n'(2) = A_0 S'\left(\frac{c'}{d'+\alpha} + \frac{k_2}{\alpha}\right) \tag{11}$$

Where the detection area *S*′ is infinite, cm^2.

The ratio of the full peak count rates of the 2th collimator to 1st collimator, *R*′(*α*), is calculated by the following Eq. (12), which depicts the relationship between the *R*′(*α*) and *α*.

$$R'(\alpha) = \frac{n'(2)}{n'(1)} = \frac{(c'+k_2)\alpha + k_2 d}{(a'+k_1)\alpha + k_1 b} \cdot \frac{b'+\alpha}{d'+\alpha} \tag{12}$$

3. Peak-to-valley ratio method

For an infinite half-space source with exponential distribution, based on Beck Eq. the full peak count rate (n'') in the in-situ spectrum can be calculated as following Eq. (13).

$$n'' = \frac{A_0 P_\gamma S_0}{2} \int_0^1 \frac{F(\cos\theta)}{(\mu_s + \alpha\cos\theta)} \cdot e^{-\mu_a h/\cos\theta} d\cos\theta \tag{13}$$

The probability (P_1) of single scattering rays emitting from soil can be expressed as the Eq. [7] (14).

$$P_1 = \int_0^{r-h/\cos\theta} k\mu_s e^{-\mu_s x} dx \cdot e^{-\mu_s(r-h/\cos\theta - x)} e^{-\mu_a h/\cos\theta} \tag{14}$$

$$= k\mu_s (r - h/\cos\theta) e^{-\mu_s(r-h/\cos\theta) - \mu_a h/\cos\theta}$$

Where *r* is the distance from radioactive unit-volume-element to detector, cm. *k* is the fraction of interacted photons scattered into the 'valley'.

Based on the Eq. (14), the net valley count rate (*C*) due to the interaction only with soil in the in-situ spectrum, can be calculated as following Eq. (15).

$$C = \frac{A_0 P_\gamma S_0 k \mu_s}{2} \int_0^1 \frac{F(\cos\theta)}{(\mu_s + \alpha\cos\theta)^2} \cdot e^{-\mu_a h/\cos\theta} d\cos\theta \tag{15}$$

Therefore, the peak-to-valley ratio ($R''(\alpha)$) can be calculated by the following Eq. (16).

$$R''(\alpha) = \frac{n''}{C} = \frac{1}{k\mu_s} \cdot \frac{\displaystyle\int_0^1 \frac{F(\cos\theta)}{(\mu_s + \alpha\cos\theta)} \cdot e^{-\mu_a h/\cos\theta} d\cos\theta}{\displaystyle\int_0^1 \frac{F(\cos\theta)}{(\mu_s + \alpha\cos\theta)^2} \cdot e^{-\mu_a h/\cos\theta} d\cos\theta} \tag{16}$$

III. Sensitivities analysis

1. Method parameters

(1) Multiple photopeak method

Given the soil density 1.6 g/cm^3 and a GMX HPGe detector, the detection efficiencies (*ε*) of 244keV and 1408keV γ-rays of ^{152}Eu are calculated by the Eq. (2), and show as following **Fig.1**, and the fitting parameters (*a*, *c*, *b* and *d*) are listed in the **table** 1 also.

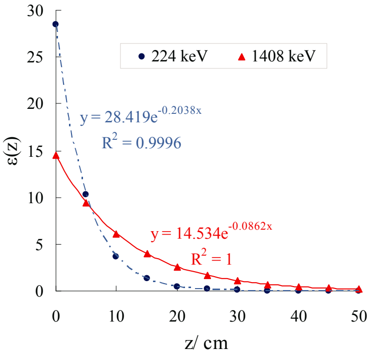

Fig.1 The detection efficiencies *ε* of 244keV and 1408keV γ-rays of ^{152}Eu.

Table 1 Parameters of Multiple photopeak method.

Parameters	*a*	*b*(cm^{-1})	*c*	*d*(cm^{-1})
Values	28.419	0.2038	14.534	0.0862

(2) Collimation or lead-plate method

As above, the detection efficiencies (ε') of 662keV γ-rays of ^{137}Cs with 30d-50mm and 90d～180d-50mm collimators are calculated using ISOCS-software, and show as following **Fig.2**, and the fitting parameters (*a*′, *c*′, *b*′, *d*′, k_1 and k_2) are listed in the **table 2** also.

Table 2 Parameters of Collimation or lead-plate method.

Par.	a'	b'(cm^{-1})	k_1	c'	d'(cm^{-1})	k_2
Val.	0.9829	0.1914	0.01074	0.9942	0.2239	0.00418

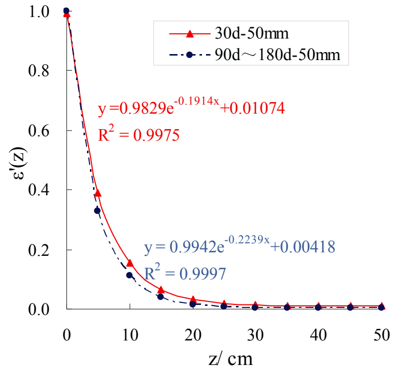

Fig.2 The detection efficiencies ε' of 30d-50mm and 90d～180d-50mm collimators.

(3) Peak-to-valley ratio method

Given F(cosθ)=1, and k an arbitrary value that couldn't affect the analysis results. The peak-to-valley ratio ($R''(\alpha)$) was calculated by the Eq. (16), and show as following **Fig.3**.

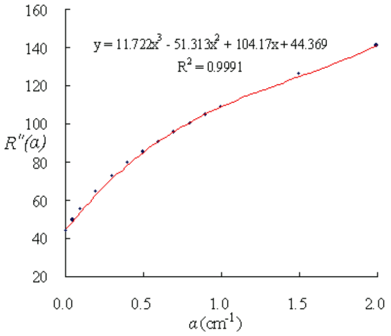

Fig.3 Relationship between $R''(\alpha)$ and α (the reciprocal of the relaxation depth)

2. Sensitivities comparison

Based on the method parameters, the values of each method-ratio above were calculated by the Eqs. (7), (12) and (16). For comparing the potential sensitivity of each method, the values of each method-ratio were normalized to 1 at a particular value of the parameter α=0 cm^{-1}. The choice of this normalization point is completely arbitrary but, for visual simplicity, was chosen to be at one extent of the realistic range. The relative relaxation-depth sensitivity graphs were given as following **Fig.4**.

As can be seen from these graphs, while 0<α<0.3cm^{-1}(corresponding to the radioactive distribution maximum depth 15~∞ cm), three methods' sensitivities are all relative high, and the sequence from the high to the low can be expressed as following: "Peak-to-valley" ≈ "Multiple photopeak" >> "Collimated or lead plate"; while α>0.3cm^{-1} (corresponding to the radioactive distribution maximum depth 0~15 cm), Peak-to-valley method shows the greatest variation in method-ratio, offering the best potential sensitivity, and Collimation or lead-plate method shows the least even neglectable variation in method-ratio, therefore the sequence of relaxation-depth sensitivity from the high to the low should be express as following: "Peak-to-valley" >> "Multiple photopeak" > "Collimated or lead plate"; In general, the sequence of relaxation-depth sensitivity from the maximum to the minimum was as following: Peak-to-valley method, Multiple photopeak method, and Collimation or lead-plate method, and the sensitivity of Collimation or lead-plate method was far less than Multiple photopeak method.

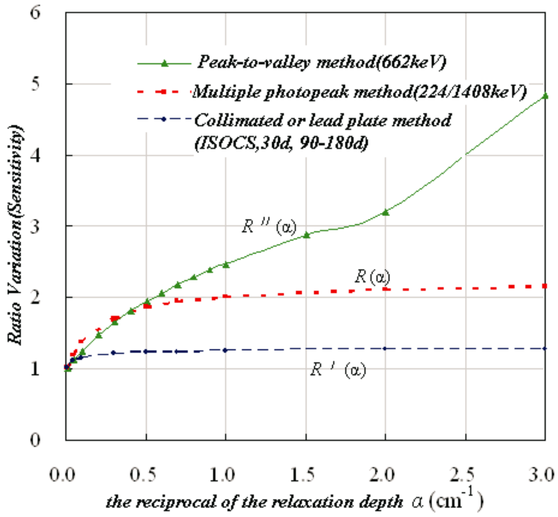

Fig.4 Sensitivities comparison of the three methods.

Comparing with the J MacDonald's results[19,20)], there are some differences in the sensitivity relationship between Peak-to-valley ratio method and Multiple photopeak method. J MacDonald's research showed that the sensitivity of Peak-to-volley ratio is less than Multiple photopeak. It is because that J MacDonald's research adopted 32keV X-rays absorbed by soil seriously, but our work employed the least energy at 224keV. In practice, 32keV X-rays is unseemliness in the field measurement due to the complicated background disturbance. In other ways, while the radionuclide distributed on the ground surface (z⟶0), the Peak-to-valley ratio would tend to become the infinity, but the ratio of Multiple photopeak method is a limited constant only. Therefore, the verdict that relaxation-depth sensitivity of Peak-to-volley ratio is higher than Multiple photopeak should be rational.

To Collimation or lead-plate method, we adopted the most prime collimator-combination (30d-50mm, and 90～180d-50mm) with the greatest variation in method-ratio. However, the changing range of the method-ratio is very small, which shows that the competence of Collimation or lead-plate method to determine the depth-distribution is very limited. And Robert's researches[21)] show that Collimation or lead-plate method only can be used to distinguish between the infinite homogeneous distribution and the surface distribution, which is according well with our results.

IV. Conclusions

By the theoretical study, the sensitivity comparison of three main methods for the depth-distribution measurement of radionuclides in soil has been performed. The experimental methods for Multiple photopeak, Peak-to-valley ratio and Collimated or lead plate are at the energy of 244keV and 1408keV γ-rays of ^{152}Eu, 662keV γ-ray of ^{137}Cs, 662keV γ-ray of ^{137}Cs, respectively, and the ISOCS was employed in Collimation or lead-plate method. Results show that the sequence of relaxation-depth sensitivity from the maximum to the minimum was as following: Peak-to-valley method, Multiple photopeak method, and Collimation or lead-plate method, and the sensitivity of Collimation or lead-plate method was far less than Multiple photopeak method.

Peak-to-valley ratio method can be applied to the arbitrary γ emitter radionuclide. Multiple photopeak one could be adopted for the radionuclide emitting at least two energy γ-rays. These two methods are more sensitive and practicable than Collimation or lead-plate method to determine the depth distribution of radionuclides in soil, and Peak-to-valley ratio method is better than Multiple photopeak method, in general. To Collimation or lead-plate method, not only it's least sensitivity, but also the potential fatal error due to radioactive unhomogeneity along the landscape-orientation, therefore it's competence to determine the depth-distribution is very limited and debatable.

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