A DEVELOPMENT FOR DETERMINING THE ACTIVITY OF RADIONUCLIDES IN THE ENVIRONMENTAL SAMPLE BY HPGE γ-SPECTROSCOPY USING ONLY ONE ABSOLUTE EFFICIENCY VALUE AND AN INTRINSIC EFFICIENCY

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Abstract: In the present work, we presented a development for determining the radio-activities in the environmental sample based on knowing only absolute (full-energy peak) efficiency value at the energy of 1460.83keV corresponding to characteristic γ-ray of ^{40}K by using a reference in separate and an intrinsic (relative) efficiency function. The prerequisite for using this method is the presence of a multi-gamma-ray emitter in the sample, with an activity allowing to measure the count rates with the statistic uncertainty of few percents. Two standard samples were used to validate the development. Gamma-rays from ^{228}Th or ^{226}Ra decay series were chosen to construct the intrinsic efficiency functions. The results obtained were in good agreement with reference data.

Keywords: *γ-spectroscopy, intrinsic efficiency curve, absolute efficiency value,RGK-1*

1. INTRODUCTION

The approach, which is widely used for activity determination of environmental samples, is related to construct the full-energy peak efficiency curve (FEPEC) of γ-spectroscopy, ε(E). the ε(E) curve can be built by experiment or simulation by the following equation:

$$\varepsilon(E_j) = \varepsilon_{ref}(E_j)C(E_j) \quad (1a)$$

The following equation gives us A_T :

$$A_T = \frac{n_j / I_{\gamma j}}{\varepsilon(E_j)} \quad (1b)$$

where n_j is the count rates, $I_{\gamma j}$ is the emission probability. The value ε_{ref} (E_j) is the absolute efficiency calculated through reference material. Meanwhile, C (Ej) = $C_s(E_j)/C_{ref}(E_j)$ is relative self-absorption correction factor and it receives the value around unit. C_s and C_{ref} are self-absorption correction factors for efficiencies of analyzed sample and reference material, respectively.

In this paper, a developed approach to calculate radionuclides in environmental samples is presented. Only the value of absolute efficiency at 1460.83 keV energy calculated based on RGK-1 and an intrinsic (relative) efficiency curve are required. The intrinsic efficiency was successfully, widely used for estimating isotopic ratios and age-dating of enriched-uranium materials by gamma-spectroscopy [1,2,3,4,5]. The intrinsic calibration was obtained based on many $n/ I_{\gamma j}$ values of the gamma-rays coming from the same isotope in the same spectrum.

For our present work, gamma-rays coming from decay series of ^{226}Ra and ^{228}Th are chosen for deducing the intrinsic calibration curve. ^{226}Ra and ^{228}Th are the daughters of ^{238}U and ^{232}Th chains, respectively. The appearance of ^{238}U and ^{232}Th dated back to the formation of the earth. Because of very long half-life, these radionuclides, as well their daughters such as ^{226}Ra and ^{228}Th, still exist in our planet and appear in most environmental samples such as soil, sediment, vegetable, fish, etc....

This technique was also described in the paper written by Felsmann et al. [6]. The measurement of the absolute efficiency at 1460.83 keV line is done by mixing reference material KCl uniformly with the investigated sample. In our work, however, reference material and investigated sample are separated.

2. METHOD AND RESULTS

2. 1. General description of the method

Assuming multi-gamma-ray emitter T in analyzed sample emits several measurable gamma-lines so that from the same gamma spectrum, one can easily calculate a series of $F_T(E_j) = nj$ ratios. By least-squares fitting dataset $F_T(E_j)$ to a suitable mathematical function, the fitted $F_T(E)$ function is obtained. Then, Eq. (1b) is re-written as:

$$F_T(E) = \varepsilon(E)\, A_T \tag{2}$$

Then, A_T can be determined by knowing only one value of full-energy peak efficiency at a certain energy E*:

$$A_T = \frac{F_T(E^*)}{\varepsilon^*} \tag{3}$$

where ε* = ε(E*), E* should be inside of energy range of nuclide T. In this paper, the 1460.83keV gamma-ray of ^{40}K in reference material RGK-1 was used to determined ε* according to Eq.(1a).

Eq.(2) expresses the relation between absolute efficiency and intrinsic efficiency. Absolute efficiency, ε(E), is common for different radionuclides in measured samples. Therefore, all γ-emitters (mono- or multi-γ emitters) in a sample have the same absolute efficiency curve and their intrinsic efficiency curve is unique for each radionuclide i.e. the relative efficiencies of isotopes in a sample are proportional to each other with a coefficient A_T.

Assuming the analyzed sample contains another radionuclide (named Y) beside T. Let's call $F_Y(E)$ the intrinsic efficiency function constructed by using gamma-lines from Y. Similarly, activity A_Y can be estimated from Eq.(3) as the case of A_T. Moreover, $F_Y(E)$ must be proportional to $F_T(E)$. Then, one has:

$$F_Y(E) = \frac{A_Y}{A_T} F_T(E) = R F_T(E) \tag{4}$$

where R is constant for a given sample, which can be derived from the measurement. It means that from Eq.(4) the activity A_Y can also be estimated from A_T and R by one of the following equations:

$$A_Y = R A_T \tag{5a}$$

$$A_Y = A_T \frac{F_Y(E)}{F_T(E)} \tag{5b}$$

After knowing F_T (E) and ε∗, all activities of γ-emitters are determined through Eqs.(3 - 5b). F_T(E) is derived from the γ-spectrum of the investigated sample, while ε∗ is calculated from the γ-spectrum of reference material. There are several reference materials approved by IAEA, from which a photo-peak can be chosen to calculate ε∗. But, to gain high precision result, some criteria should be satisfied:

- The peak corresponding to E∗ should not be interfered by other peaks in the same spectra.
- True coincidence summing (TCS) is not necessary to be performed for the chosen peak.
- The activity of radioisotope which emits gamma-line corresponding to the chosen peak is very high compared to the natural background so that the uncertainty from the background can be reduced.
- Relative self-absorption correction factor C(Ej) in Eq.(1a) comes close to 1 with increasing the energy. For many environmental samples, C(Ej) could be approximated with unity for the γ-ray with energy above 1 MeV.

Based on these criteria, RGK-1 could be a good candidate because this reference material contains mainly ^{40}K radionuclide[7]. And ^{40}K emits only one measurable gamma ray with an energy of 1460.83keV [8]. In this case, E∗ is equal to 1460.83keV.

2. 2. Validation Experiment

Two samples were prepared based on reference materials [7, 9, 10], called Sample-1 and Sample-2. Sample-1 is directly RGTh-1 materials. Sample-2 is secondary reference material based on the mixture of RGU-1 (12.00±0.01g), RGTh-1 (12.10±0.01g) and RGK-1 (12.60±0.01g), SiO_2(57.35±0.01g), $CaCO_3$ (57.35±0.01g), MgO (19.10±0.01g) and TiO (9.50±0.01g). The activity concentrations of the three radionuclides (^{226}Ra, ^{228}Th and ^{228}Ra) in this sample were computed from the available isotopic data [9, 10] and shown in Table 3.

These samples were contained in a polyethylene cylindrical box of 7.6 cm × 3 cm (diameter × height). Each sample has a mass of 180 g. A commercial press was used to ensure all sample having the same density. They all were sealed for more than 3 weeks to establish secular equilibrium [11] before measurements.

Another polyethylene cylindrical box filled with RGK-1 material, labeled as Sample-Ref was considered as reference material to calculate ε_{ref}. The dimension and mass of Sample-Ref are exactly the same as Sample-1 and Sample-2.

2. 3. Data analysis and Results

Since peaks coming from gamma-rays for ^{228}Th and ^{226}Ra decay series appeared dominantly in the gamma spectra of Sample-1 and Sample-2, it would be convenient to construct intrinsic efficiency function based on these gamma-rays. Activity concentrations of radionuclides were derived from the measured gamma-spectra using the method described in section 2 as follows:

- Calculate ε∗ and relative self-absorption factor at E∗.
- Construct F_T(E) by fitting a suitable mathematical model to $n_j/I_{\gamma j}$ ratios.
- Calculate A_T and other radio-activities by using Eqs.(3 - 5b).

2. 3. 1. Calculate absolute efficiency and relative self-absorption factor at 1460.83 keV

Only one gamma-ray with the energy of 1460.83 keV is emitted by ^{40}K [8]. In the environmental sample, the 1460.83 keV peak is interfered by 1459.138 keV peak of ^{228}Ac. But in the case of RGK-1, this interference is ignorable [7]. The obtained efficiency at 1460.83 keV is 0.532 ± 0.003 (Fig. 9). This value will be used in Eq.(2a) to calculate absolute efficiency,ε*, for Sample-1, Sample-2.

Since the densities of Sample-1, Sample-2 and Sample-Ref are equal to each other (1.3 g/cm^3). The relative self-absorption correction factors for (Sample-1, Sample-Ref) pair and (Sample-2, Sample-Ref) pair are safely considered as a unity. However, to check this hypothesis, another experiment was performed using a transmission method proposed by Cutshall et al. [12]. The C(1460.83 keV) = 1.001 ± 0.001 was found between Sample-1 and Sample-Ref, while = 1.003 ± 0.001 was for (Sample-2, Sample-Ref) pair (Fig.10).

2. 3. 2. Gamma-rays of ^{226}Ra and ^{228}Th decay series for constructing $F_T(E)$

In general, the gamma-rays emitted directly by ^{228}Th cannot be measured by gamma spectroscopy because of low emission probability. ^{226}Ra has a detectable gamma-ray with the energy of 186.211 keV, but this peak is totally overlapped by 185.712 keV peak of ^{235}U. Fortunately, the short-lived daughters of these isotopes emit intense gamma-rays measurable by the gamma spectroscopy. By sealing sample container long enough for establishing secular equilibrium between the short-lived daughters and their parent, we obtain:

$$A_{Ra226} = A_{Pb214} = A_{Bi214} \tag{6a}$$

$$A_{Th228} = A_{Pb212} = A_{Bi212} = \frac{A_{Tl208}}{p} \tag{6b}$$

where A_{Ra226}, A_{Th228}, ..., denote the corresponding activities at the time of measurement, while p = 0.3594 is the decay branching probability of the decay of ^{212}Bi to ^{208}Tl.

Assuming $F_{Ra226}(E)$, $F_{Th228}(E)$ is the intrinsic efficiency calibration curves corresponding to ^{226}Ra, ^{228}Th, respectively. Because 1460.83 keV of ^{40}K in RGK-1 reference material is chosen to calculate ε*, the peaks used to build $F_{Ra226}(E)$, $F_{Th228}(E)$ have to distribute in a range of energy which 1460.83 keV is located within. Table 1 listed the energies and their emission probability which were used in this paper.

Table 1: The energies and emission probabilities of the γ-peaks used in this work [8].

Energy (keV)	Emission probability	Emitter
^{226}Ra group		
295.224	19.3 (2)	^{214}Pb
351.932	37.6 (4)	^{214}Pb
609.312	46.1 (5)	^{214}Bi
1120.287	15.1 (2)	^{214}Bi
1764.494	15.4 (2)	^{214}Bi
2204.21	5.08 (4)	^{214}Bi
^{228}Th group		
238.632	43.3 (2)	^{212}Pb
583.191	84.5 (7)	^{208}Tl
860.564	12.42 (10)	^{208}Tl
1620.50	1.49 (3)	^{212}Bi
2614.533	99	^{208}Tl
^{228}Ra group		
338.321	11.27 (19)	^{228}Ac
794.947	4.25 (7)	^{228}Ac
911.204	25.8 (4)	^{228}Ac
968.971	15.8 (3)	^{228}Ac

2. 3. 3. Calculate the activity of radionuclides in Sample-1

2. 3. 3. 1. ^{228}Th

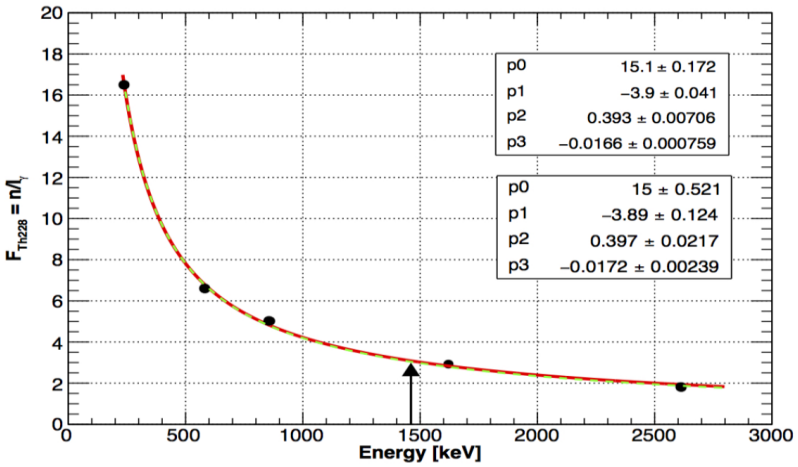

Figure 2: The intrinsic efficiency curve of ^{228}Th in Sample-1. The circles present the experimental data. Meanwhile, two lines are fit curves: the red one is for the case of using 1620.74 keV, and the dash, the green one is corresponding to the case of not using 1620.74 keV. The arrow shows the position of 1460.83 keV. The error bars are not seen because of being smaller than the size of symbols.

The $F_{Th228}(E)$ was obtained by fitting the Eq. (7) to the $n_j/I_{\gamma j}$ ratios at 238.832 keV of ^{212}Pb, 1620.74 keV of ^{212}Bi and 583.191, 860.564, 2614.533 keV of ^{208}Tl. Since the emission probability of 1620.74 keV is rather small in comparison with other gamma-rays (see Table 1), and the distance between 860.564 and 2614.533 keV is large, so it is worth testing that how well $F_{Th228}(E)$ behaves with or without using $n_j/I_{\gamma j}$ ratio at 1620.74 keV in the curve fitting. The results of this check have meanings in a reality where 1620.74 keV peak is not always seen in the gamma spectrum of the environmental sample. After $F_{Th228}(E)$ is determined, the result $A_{Th228}=A_{Pb212}=A_{Bi212}=A_{Tl208}/p$ is found and shown in Table 2. The A_{Th228} with the use of 1620.74 keV was closer to reference data. The small box on the left of Fig. 2 showed the fit parameters: upper one is for the case of using 1620.74 keV, the lower one is contrary.

$$F_T(E) = Exp\left(p_0 + p_1 ln(E) + p_2 ln(E)^2 + p_3 ln(E)^3\right) \quad (7)$$

Table 2: Results for Sample-1. All the results were expressed with 95% confidence intervals.

Radionuclide	Activity (this work) Bq/Kg	Activity (Ref. [16]) Bq/Kg	Deviation (%)
$^{228}Th = {}^{212}Pb = {}^{212}Bi = {}^{208}Tl/p$	3236 ± 76	3250 ± 88	-0.43
$^{228}Ra = {}^{228}Ac$	3268 ± 77	3250 ± 88	0.55
$^{226}Ra = {}^{214}Pb = {}^{214}Bi$	76.00 ± 1.78	78 ± 6	-1.6

2. 3. 3. 1. ^{228}Ra and ^{226}Ra

The activity of ^{228}Ra was derived through the gamma-rays of ^{228}Ac due to secular equilibrium. The four measurable gamma-rays of ^{228}Ac were 338.32, 794.947, 911.204, 968.971 keV. It is easily seen that 1460.83 keV is far away out of this energy range. Hence, if fitting directly equation 7 to n/Iγ ratios at these energies, the value of $F_{Ra228}(E)$ at 1460.83 keV would fluctuate very strongly, and give more uncertainty. In this case, Eq. (4) was used to overcome this difficulty. For more clear, the following function is dedicated to extracting $F_{Ra228}(E)$:

$$F_{Ra228}(E) = p_4 Exp(15.1 - 3.9ln(E) + 0.393ln(E)^2 - 0.0166ln(E)^3) \quad (8)$$

where all the numbers in Eq.(8) were corresponding to fit parameters p0, p1, p2, p3 of $F_{Th228}(E)$ (see the upper box on Fig. 2). Looking at the box on the upper right of Fig.3, the uncertainties of p0, p1, p2, p3 are zero. This implied that these values were fixed in the fit. The parameters p4 is equal to R in Eq.(4), hence from Eq.(5a), A_{Ra228} is determined as :

$$A_{Ra228} = p_4 A_{Th228} \quad (9)$$

$F_{Ra226}(E)$ and A_{Ra226} in Sample-1 would be determined exactly as the same way as

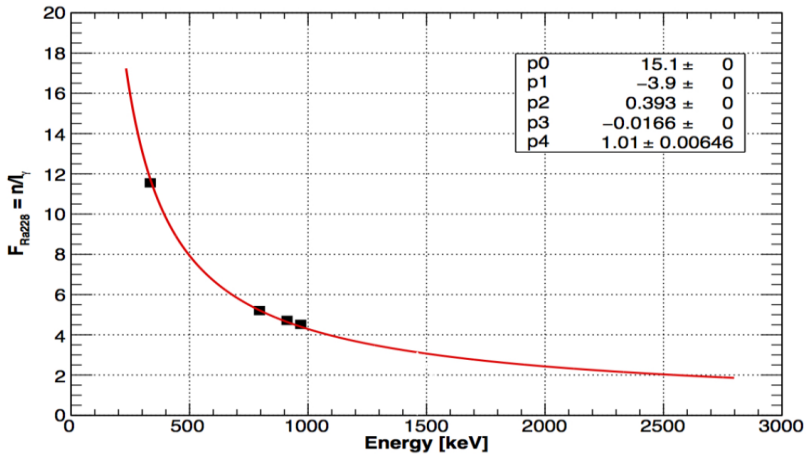

Figure 3: The intrinsic efficiency curve of ^{228}Ra in Sample-1 obtained by using Eq. (8) which contains $F_{Th228}(E)$. The only free parameter in the fit is p4 shown in the box. The black square symbol presents the experimental data. The red line is the fit curve. The error bars are not seen because of being smaller than the size of symbols.

$F_{Ra228}(E)$ and A_{Ra228}, but at 351.931, 609.312 keV. Fig.4 shows the intrinsic efficiency curve for ^{226}Ra in Sample-1. The results for A_{Ra226} and A_{Ra228} in Sample-1 are listed in Table 2. Compared to reference data, our results gave a good agreement.

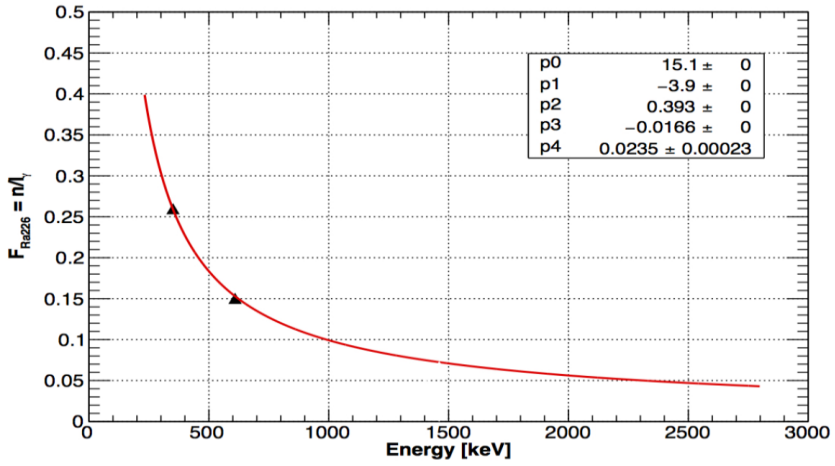

Figure 4: The intrinsic efficiency curve of ^{226}Ra in Sample-1 obtained in the same way as $F_{Ra228}(E)$. The black square symbol presents the experimental data. The red line is the fit curve. The error bars are not seen because of being smaller than the size of symbols.

2. 3. 4. Calculate the activity of radionuclides in Sample-2

In gamma-spectrum of Sample-2, only ^{226}Ra had enough number of photo- peaks which

had high statistic for building $F_T(E)$ function. Fig. 5 presents the $F_{Ra226}(E)$ curve. The red smooth line is obtained by fitting the Eq. (7) to n/Iγ ratios at 295.224, 351.932 keV of ^{214}Pb and 609.312, 1120.287, 1764.494, 2204.21 keV of ^{214}Bi . The activities A_{Th228} and A_{Ra228} were determined through $F_{Ra226}(E)$ by using Eq.(5b):

$$A_{Th228} = \frac{n_{583keV}/I_{\gamma 583keV}}{0.3594 F_{Ra226}(583keV)} A_{Ra226} \quad (10a)$$

$$A_{Ra228} = \frac{n_{911keV}/I_{\gamma 911keV}}{F_{Ra226}(911keV)} A_{Ra226} \quad (10b)$$

Table 3 listed the results for A_{Th228}, A_{Ra228} and A_{Ra226} in Sample-2. The results were in good agreement with reference data.

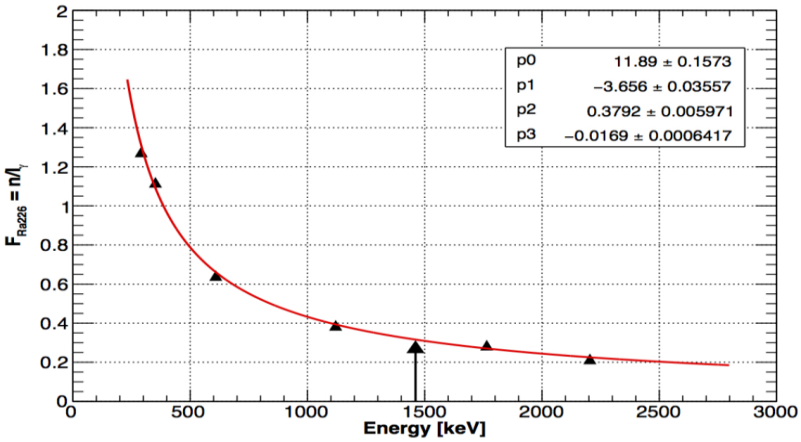

Figure 5: The intrinsic efficiency curve of ^{226}Ra in Sample-2. The black symbol presents the experimental data. The red line is the fit curve. The arrow shows the position of 1460.83 keV. The error bars are not seen because of being smaller than the size of triangles.

Table 2: Results for Sample-1. All the results were expressed with 95% confidence intervals.

Radionuclide	Activity (this work) Bq/Kg	Activity (Ref. data) Bq/Kg	Deviation (%)
$^{228}Th = {}^{212}Pb = {}^{212}Bi = {}^{208}Tl/p$	216.2 ± 8.4	218.5 ± 6.0	-1.05
$^{228}Ra = {}^{228}Ac$	218.2 ± 8.5	218.5 ± 6.0	-0.14
$^{226}Ra = {}^{214}Pb = {}^{214}Bi$	329.8 ± 6.4	329.3 ± 2.0	0.15

3. CONCLUDING REMARKS

A development for determining the radio-activities in environmental samples using only one value of detector full-energy peak efficiency was presented. The ”holy grail” of this method is to build an intrinsic efficiency calibration from experimental n/Iγ ratios of the same radionuclide in the same spectrum. This is also the limitation of this method.

The intrinsic efficiency function will be improved by constructing from gamma lines of all radionuclides in the measured sample. In this paper, gamma-rays from ^{228}Th or ^{226}Ra series were used for building intrinsic efficiency function, $F_T(E)$. Some gamma-lines can encounter true coincidence summing effect. Correction factors can be determined by Geant4 simulation. In environmental measurements, however, acceptable counting the uncertainties are usually high such that coincident summing corrections might be ignored [13]. At present, only activities of

radionuclides which emit measurable gamma-line with energy from 238keV-3000keV can be determined. In future work, the $F_T(E)$ function will be extended for energy below 238 keV.

IAEA-RGK-1 was chosen as a perfect candidate for determining ε* because of fulfilling criteria mentioned in Section.2. The accuracy of results derived from this method depends on the precision of the value of intrinsic efficiency function and the absolute efficiency at the energy of 1460.84keV corresponding to ^{40}K's γ-ray. IAEA-RGK-1 and investigated samples are separate in our work. This differs from the work proposed by Felsmann et al. [6], where reference material KCl was blended uniformly into investigated samples.

The results obtained in our work were confirmed by environmental reference materials (Sample-1, Sample-2). It can be concluded that the method described in this paper can be a reliable alternative for radioactivity determination of environmental samples.

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PHÁT TRIỂN PHƯƠNG PHÁP TÍNH TOÁN HOẠT ĐỘ CỦA NHÂN PHÓNG XẠ TRONG MẪU MÔI TRƯỜNG BẰNG PHỔ KẾ γ-HPGE CHỈ SỬ DỤNG MỘT GIÁ TRỊ HIỆU SUẤT TUYỆT ĐỐI VÀ ĐƯỜNG CHUẨN TRONG

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Tóm Tắt: Trong báo cáo này, chúng tôi trình bày một sự phát triển để xác định các hoạt độ phóng xạ trong mẫu môi trường dựa trên việc chỉ biết một giá trị hiệu suất tuyệt đối ở năng lượng 1460.83keV tương ứng với tia gamma đặc trưng 40K bằng cách sử dụng mẫu chuẩn riêng biệt và đường chuẩn trong. Điều kiện tiên quyết để sử dụng phương pháp này là sự hiện diện của đồng vị phát nhiều tia gamma trong mẫu, với hoạt động cho phép đo tốc độ đếm với sai số thống kê một vài phần trăm. Hai mẫu chuẩn đã được sử dụng để đánh giá sự phát triển. Các tia gamma từ chuỗi phân rã 228Th hoặc 226Ra đã được chọn để xây dựng các hàm chuẩn trong. Các kết quả thu được phù hợp tốt với số liệu tham khảo.

Từ khoá: Phổ kế *γ, đường chuẩn trong, Hiệu suất ghi tuyệt đối,,RGK-1*