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A Suggested Modification of the Fundamental Parameter Method: A Case Study to Calculate the Optimum Absolute Intensity of 1001.03 keV Gamma-Ray Transition

Ashraf E.M. Khater[1] and Yasser Y. Ebaid[2]

[1] *Nuclear Safeguards and Physical Protection Department, Nuclear and Radiological Regulatory Authority, Cairo, Egypt*

[2]*Physics Department, Faculty of Science, Fayoum University, Fayoum, Egypt*

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The fundamental parameter method (FPM) is an analytical approach for intrinsic calibration of gamma-ray spectrometer using fundamental nuclear and atomic parameters such as gamma-ray branching intensity, half-life time, isotopic ratio and concentration ratio. The main advantage of this approach is the wide range of its applications in gamma-ray efficiency calibration, nuclear safeguards (nuclear materials measurement and isotopic ratios) and others. In this work, the calculation of the relative efficiency (RE: photopeak count rate divided by branching ratio) was modified based on the relative intensity concept of $^{-226}$Ra in equilibrium with ^{222}Rn decay products (^{214}Bi-^{214}Pb). The modified FPM was applied to reevaluate the absolute intensity (I%) of gamma-ray transition of ^{234m}Pa at 1001.03 keV using certified uranium ore samples, ^{226}Ra point source and gamma-ray spectrometers based on hyper pure germanium detector. The newly confirmed I % of 1001.03 keV is 1.0164±0.0636.

Keywords: Fundamental parameter method/ 1001.03 keV/ Gamma-ray transition/ ^{238}U/ 1001.03 keV

Introduction

The fundamental parameter method was suggested by Eberle et al. as a direct application of peak ratio concept and intrinsic calibration of gamma-ray spectrometer using fundamental nuclear and atomic parameter such as gamma-ray branching intensity (I%), half-life time, isotopic ratio and concentration ratio. This approach was applied for the measurement of the isotopic ratios of different nuclear materials in the nuclear safeguard field. It was also used to confirm the squallier equilibrium with natural radionuclides (e.g. ^{238}U and ^{232}Th) series decay products [1]. The RE concept was proposed to use the spectrum of the sample itself to estimate the variation of detector efficiency as a function of energy where RE is the ratio of photopeak count rate to I% of the same gamma-ray transition energy.

The accurate determinations of ^{238}U and other U isotopes (^{235}U and ^{234}U) are the backbone of different ^{238}U series disequilibrium applications and nuclear safeguards [2-5]. Gamma-ray spectrometer is one of the widely used non-destructive analytical techniques for determination of U activity concentration and isotopic ratio. For ^{238}U, the well resolved gamma-ray transition of ^{234m}Pa (granddaughter of ^{238}U) at 1001.03 keV is preferably used because of its minimal self-attenuation and spectral interferences [6-13]. The accurate measurement of gamma-ray emitters lied

Corresponding author: khater_ashraf@yahoo.com
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on the accuracy of I% value (also called branching ratio, emission probability and f-value). Earlier, Coursol et al, reported an intensity value of 0.59% for the 1001.03 keV that was used for U gamma-ray evaluation for a while [14]. Nodaway, the commonly used I% value is 0.847±0.008% [15].
Among all the ruling factors affecting the accuracy of gamma-ray spectrometry, I % relatively overcomes the significance of other parameters. Over decades, various studies have been performed in order to accurately evaluate I% of 1001.03 keV gamma-ray transition that varies widely from 0.59% up to 1.12% [16-35]. Furthermore, recent studies have suggested that there is a noticeable under estimation of the currently used I% of 1001.03 keV gamma-ray transition. The most recent published values of I% were 1.037±0.052 and 1.067±0.084 using both of the absolute and relative efficiency calibration concepts, respectively [36-37].
This study aims at suggesting modification on the FPM, in addition to optimizing and scrutinizing I% value of the 1001.03 keV gamma-ray transition of the ^{234m}Pa using MFPM.

A modified fundamental parameter method

The activity ratios of ^{238}U series members (where the activity ratios amongst them are approximately unity) using gamma-ray spectrometer and energy transitions of ^{234}Th, ^{234m}Pa, ^{226}Ra, ^{214}Pb and ^{214}Bi, along with the status of U series secular equilibrium can be confirmed using the fundamental parameter method (FPM) that was described in details by Eberle [1]. It does not need standard sources, and it depends on physical parameters such as the absolute intensity and the half-life. The relative efficiency (RE) concept facilitates the generation of a RE curve from the sample itself regardless of their geometry, matrix or activity concentration, taking into consideration some spectrometric vital factors such as intrinsic efficiency of the detector, counting geometry and attenuation in the sample matrix [30]. The principle equation for activity concentration calculation is;

$$A = \frac{C}{\varepsilon \times I} \qquad (1\text{-a})$$

$$\varepsilon = \frac{C}{A \times I} \qquad (1\text{-b})$$

Where

ε : absolute full energy photopeak efficiency,
C : count rate (counts/sec) of 1001.03 keV photopeak,
A : activity concentration of ^{238}U in the sample, Becquerel (Bq),
I: absolute intensity % of 1001.03 keV gamma-ray transition.

Secular equilibrium of natural radionuclides series such as ^{238}U and ^{232}Th series can be confirmed using the activity ratios between series radionuclides utilizing the fundamental parameter method (FPM) and the relative efficiency (RE) concept that is given by;

$$\text{Realtive Efficiency, RE }(E_i) = \frac{\text{net count rate in the photopeak }(E_i), (\text{cps})}{\text{Absolute intensity }(E_i), (\%)} \qquad (2)$$

The details of the FPM were described in another publication by Eberle et al. [1] where the activity ratio of series radionuclides such as ^{234}Th, ^{234m}Pa, ^{226}Ra, ^{214}Pb and ^{214}Bi within ^{238}U series can be given by equations (3);

$$\frac{A_i}{A_j} = \frac{C\,(E_i)}{C\,(E_j)} \cdot \frac{\varepsilon(E_j)}{\varepsilon\,(E_i)} \cdot \frac{I\,(E_j)}{I(E_i)} \qquad (3)$$

$$\frac{\varepsilon(E_j)}{\varepsilon\,(E_i)} = \frac{RE\,(E_j)}{RE\,(E_i)} \qquad (4)$$

$$\frac{A_i}{A_j} = \frac{C\,(E_i)}{C\,(E_j)} \cdot \frac{RE(E_j)}{RE\,(E_i)} \cdot \frac{I\,(E_j)}{I(E_i)} \qquad (5)$$

Where;

$\frac{A_i}{A_j}$: The activity ratio of isotopes i and j with the gamma-ray transitions E_i and E_j

$\frac{C\,(E_i)}{C\,(E_j)}$: The net count rate (cps) ratio of photopeak of the gamma-ray transitions E_i and E_j

$\frac{\varepsilon(E_j)}{\varepsilon\,(E_i)}$: the absolute full energy photopeak efficiency ratio of the gamma-ray transitions E_j and E_i

$\frac{RE(E_j)}{RE\,(E_i)}$: The relative efficiency ratio of the gamma-ray transitions E_j and E_i

$\frac{I\,(E_j)}{I(E_i)}$: The absolute intensity ratio of the gamma-ray transitions E_j and E_i

In the modified fundamental parameter method (MFPM), the RE of different gamma-ray transitions (242-2447.86 keV) of ^{214}Pb-^{214}Bi, in all

RGU-1 voluminous samples as well as ^{226}Ra point source were calculated using the following equations instead of equation (2);

$$Reltaive\ intenisty, RI(E_i) = \frac{\text{net count in the photopeak } (E_i)}{\text{net count in the photopeak } (609\ keV)} \quad (6)$$

$$\text{Realtive Efficiency, RE } (E_i) = \frac{Calculated\ RI(E_i)}{Reference\ RI\ (E_i)} \quad (7)$$

Reference RI (E_i) for Pb-214-Bi-214 is given in other publications [15, 38, 39].

Experimental work

Aliquots of the RGU-1 reference sample [40] of different volumes were packed into standard volume cylindrical polyethylene containers. The samples where tightly sealed and put aside for 28 days in order to acquire secular equilibrium between ^{226}Ra and ^{222}R [41]. Gamma ray spectrometers based on extended range HPGe detectors were exploited. They were fully described in a recent work [36].

Samples (RGU-1) were measured for long times (up to one week) in order to insure minimal counting uncertainty in peak area calculation. Four sets of gamma-ray spectrometric measurements were performed for different samples' geometries (30, 40, 56 and 112 cc).

Radium-226-point source was measured in nine different systematical positions in circular plane (with 15 cm diameter) coaxial with and 25 cm away from the detector's end cap.

Most of the intense energy transition photopeaks of ^{214}Pb-^{214}Bi, beginning form the energy transition 242 keV (7.43%) of the ^{214}Pb and ending with 2447.86 (1.548%) of ^{214}Bi were exploited and achieved counting errors between <0.1% and 0.2%. However, the achieved counting errors for the 1001.03 keV were less than 1%. The RE of each gamma-ray transition was calculated using equation (7), and then the REs were together exploited to generate RE's polynomial fitting curves of 4th or 5th degree for each RGU-1 sample as well as for each ^{226}Ra point source measurement Fig. (1). Then the polynomial fitting functions were used to calculate the RE_p values of 1001.03 keV gamma-ray transition of ^{234m}Pa and other gamma-ray transitions (609.31, 934.06 and 1120.29 keV).

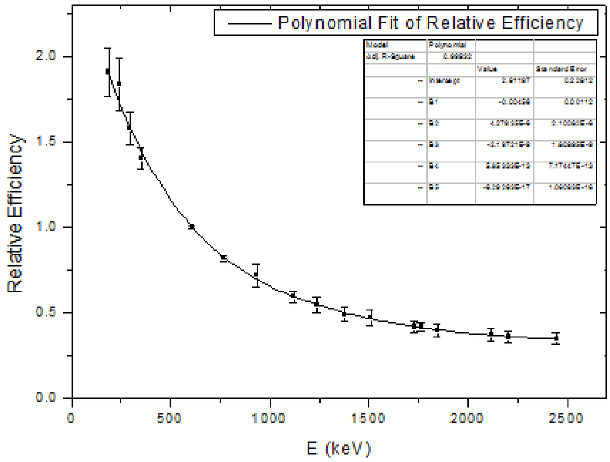
Additionally, I (E_j) of 1001.03 keV (E_j) gamma-ray transition was calculated using equation (5) and the parameters [RE_i, C (E_i) and I (E_i)] of the three gamma-ray transitions of ^{214}Bi, the most intense gamma-ray transitions are 609.31 keV (45.49 %), and the closest gamma-ray transitions to 1001.03 keV is 934.06 keV (2.89 %), and 1120.29 keV (14.91 %). These three calculated values of I % will be referred to as different MFPM modes throughout the manuscript.

The details on the sources of uncertainty and their calculations were given in a previous work [36].

Results and Discussion

The accurate determination of ^{238}U using ^{234m}Pa gamma-ray transition of 1001.03 keV depends on different parameters such as full energy photopeak efficiency (ε) and absolute intensity (I%). One of the most critical parameters is I% that has been evaluated using different analytical techniques and varied from 0.59±0.1% up to 1.12±0.067%) [16-35]. The most recent published I% value was 1.037 that was about 20% higher than the commonly used value 0.847± 0.008 [15, 36, 41].

The descriptive statistics (mean, standard deviation and range) of the absolute intensity of 1001.026 keV energy transition of the ^{234m}Pa using different modes of MFPM and different sample's geometry are given in Tables (1 and 2). Their frequency distribution is shown in Fig. (2). There is a noticeable consistency throughout the repeated



calculations

Fig. (1): The relative efficiency curve as a function of energy (keV) for the different gamma-ray transitions of 214Pb-214Bi (242-2447 keV)

The absolute intensity value (I %) of ^{234m}Pa gamma-ray transition at 1001.03 keV using the current approach (MFPM), has resulted in an optimum I% value of 1.0164± 0.0636 (0.8554-1.2234). The frequency distribution of these data is shown in Fig. (2).

Finally, the overall average ±SD (range), of the present work as well as that of the previously published works [36,37] (I % of ^{234m}Pa gamma-ray transition at 1001.03 keV)using diverse analytical approaches (AEM, FPM and MFPM), gamma-ray spectrometers, sample-detector's geometries and samples' geometries was 1.0385±0.0771 (0.8554-1.3024).

The compilation of an extensive experimental data (736 points) [36, 37] on the re-evaluation of I% of 1001.03 keV gamma-ray transition using different analytical approaches (AEM, FPM and MFPM), voluminous sources (uranium ore and granite), ^{226}Ra point source, samples' geometries, sample-to-detectors' geometries, samples' physical forms (solid and solution) and gamma ray spectrometers resulted in a new value of 1.0385± 0.0771 %.

Table (1): The absolute intensity of 1001.026 keV energy transition of the ^{234m}Pa using different modes of modified fundamental parameter method (MFPM) for its different modes

	MFPM modes	609.31 keV	934.06 keV	1120.29 keV	average
Gamma- ray Spectrometer	1	0.9890± 0.0741[+] (0.8554-1.1458)	1.0056± 0.0354 (0.9160-1.0708)	0.9737± 0.0273 (0.8991-1.0485)	1.0014 ±0.0586 (0.8554-1.2185)
	2	1.0527± 0.0620 (0.9611-1.2234)	1.0552 0.0634 (0.9651-1.2185)	1.0238 0.0619 (0.9197-1.1794)	1.0439 0.0636 (0.9197-1.2234)
	Voluminous sample (RGU-1)	1.0801± 0.0548 (0.9724-1.2234)	1.0328± 0.0548 (0.9444-1.2047)	1.0136± 0.0585 (0.9377-1.1794)	1.0422± 0.0621 (0.9377-1.2234)
	Point source, ^{226}Ra	0.9775± 0.0590 (0.8554-1.1647)	1.0219± 0.0569 (0.9121-1.2185)	0.9840± 0.0438 (0.8991-1.1388)	1.0048± 0.0610 (0.8554-1.2185)
Average		1.01633±0.0758 (0.8554-1.2234)	1.0269±0.0549 (0.9160-1.2185)	0.9952±0.0516 (0.8991-1.1794)	1.0164±0.0636 (0.8554-1.2234)

Table (2): The absolute intensity of 1001.026 keV energy transition of the ^{234m}Pa using different modes of modified fundamental parameter method (MFPM) for different sample's geometries

	Geometry	112 cc[+]	56 cc	40 cc	30 cc
Gamma- ray Spectrometer	1	0.9776 ±0.0426 (0.9377-1.0948)	1.0062± 0.0494 (0.9528-1.1657)	1.0026 ±0.0551 (0.9182-1.1458)	0.9814± 0.0578 (0.8554-1.0737)
	2	1.0088± 0.0382 (0.9245-1.0884)	1.0380± 0.0626 (0.9197-1.1657)	1.0828± 0.0752 (0.9615-1.2234)	1.0381± 0.0448 (0.9448-1.1137)
Relative Efficiency	Voluminous sample (RGU-1)	1.0077± 0.0447 (0.9377-1.0948)	1.0387± 0.0568 (0.9528-1.1657)	1.0702± 0.0724 (0.9702-1.2234)	1.0507± 0.0447 (0.9824-1.1137)
	Point source, ^{226}Ra	0.9730± 0.0374 (0.9122-1.0948)	1.0010± 0.0494 (0.9205-1.1361)	1.0026± 0.0551 (0.9182-1.1458)	0.9814± 0.0578 (0.8554-1.0737)
	Average	0.9869± 0.0437 0.9122-1.0948)	1.0161± 0.0564 (0.9197-1.1657)	1.0347± 0.0748 (0.9182-1.2234)	0.9910± 0.0578 (0.8554-1.0878)
	Average [x]	1.0164±0.0636 (0.8554-1.2234)			

[+]Mean ± standard deviation (range) [x] average of all data (357 data points)

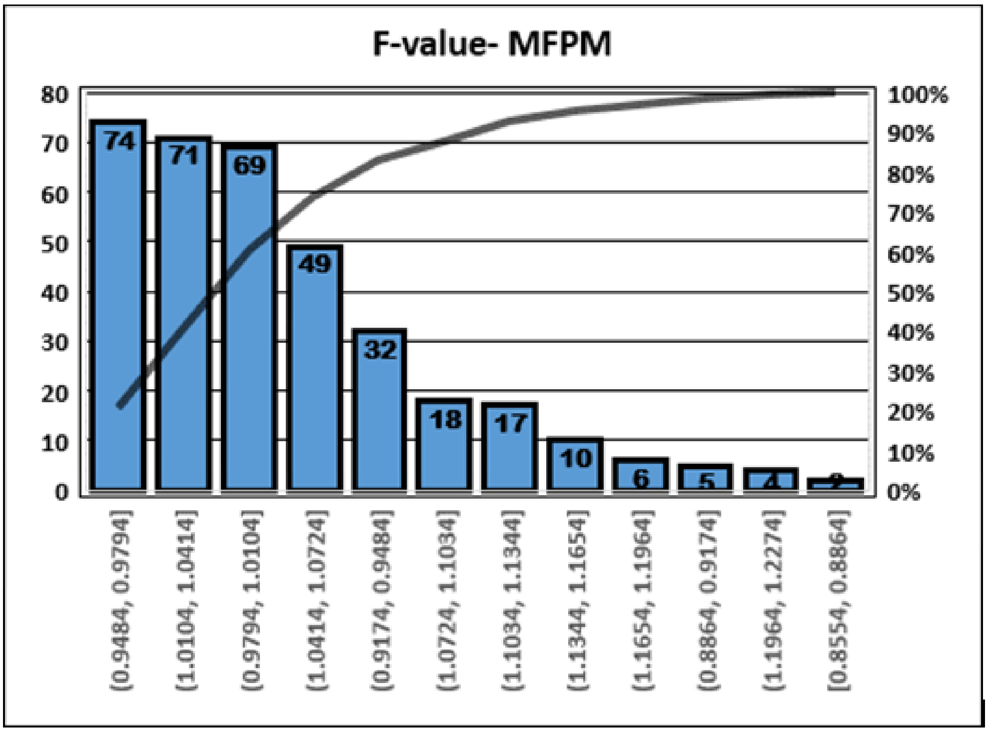


Fig. (2): Frequency distribution of the absolute intensity of 1001.03 keV energy transition of the ^{234m}Pa using modified fundamental parameter (MFPM) method

Conclusion

Based on several published experimental as well as evolutional works, the commonly applied absolute intensity (I %) of ^{234m}Pa 1001.03 gamma-ray transition is about 0.847±0.008% with a wide range from 0.59 to 1.12. The recent published I% [36] revealed a new value of 1.037±0.052%, using absolute efficiency method (AEM), that was about 22% higher than the commonly used value (0.847±0.008%). While, using a relative method called fundamental parameter method (FPM) revealed I% value of 1.0666 ± 0.0834 % with an average relative bias of 26% from the commonly used value. To confirm this newly revealed value, this study re-evaluated the I% of 1001.03 keV using the modified FPM (MFPM) and revealed the value of 1.0164 ± 0.0636% that confirmed the previous findings.

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