An approach to evaluate the efficiency of γ-ray detectors to determine the radioactivity in environmental samples

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Abstract: This work provides an approach to determine the efficiency of γ-ray detectors with a good accuracy in order to determine the concentrations of either naturally occurring or artificially prepared radionuclides. This approach is based on the efficiency transfer formula (ET), the effective solid angles, the self- absorptions of the source matrix, the attenuation by the source container and the detector housing materials on the detector efficiency. The experimental calibration process was done using radioactive (Cylindrical & Marinelli) sources, in different dimensions, that contain aqueous ^{152}Eu radionuclide. The comparison point to a fine agreement between the experimental measured and calculated efficiencies for the (NaI & HPGe) detectors using volumetric radioactive sources.

Key words: γ-ray detector efficiency, efficiency transfer, effective solid angles, radioactive source

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1 Introduction

The activity measurements of artificiality and naturally occurring radionuclides have a great impact on our lives and on our environment. These measurements require a γ-spectrometer of well-known efficiencies over a wide range of photon energies [1, 2].

The quality of the results of gamma spectrometry measurement depends directly on the efficiency accuracy for specific measurement conditions [3]. Experimental efficiency calibration is restricted to several measurement geometries in some limits and cannot be applied directly to all measurement configurations. Consequently, an alternative possibility to compute the efficiencies is highly desirable. The efficiency transfer (ET) method offers a practical and convenient solution to the problems arising from the facilities available in radiation [4]. The ET method can be used to calculate the efficiency of the detector under measuring conditions that are different from those of calibration [5] on the basis of the variation in the geometrical parameters of the source detector arrangement. This is based on the assumption that the detector efficiency $\varepsilon(E,P_{\rm o})$ at a reference position $P_{\rm o}$ is the combination of the detector intrinsic efficiency $\varepsilon_{\rm i}(E)$ depending on energy E and geometrical factors, where $\varepsilon(E,P_{\rm o})$ is given by the following equation:

$$\varepsilon(E,P_{\rm o})=\varepsilon_{\rm i}(E)\cdot\Omega(E,P_{\rm o}), \quad (1)$$

where $\Omega(E,P_{\rm o})$ is the effective geometrical solid angle of the detector to the reference source. In fact, these geometrical factors include the attenuation effects of the materials between the source and the detector, besides the self-absorption factor, if the study deals with radioactive volumetric sources. Consequently, for any point P, the efficiency can be expressed as a function of well-known reference efficiency at the same energy E and is given by:

$$\varepsilon(E,P)=\varepsilon(E,P_{\rm o})\cdot\frac{\Omega(E,P)}{\Omega(E,P_{\rm o})}, \quad (2)$$

where $\Omega(E,P)$ is the effective geometrical solid angle between the source under treatment and the detector, so the transfer coefficient $T(E,P)$ is defined as:

$$T(E,P)=\frac{\Omega(E,P)}{\Omega(E,P_{\rm o})}. \quad (3)$$

This transfer formula is generalized to volume sources by computing the relevant solid angle and absorbing factors. The main advantage of the ET method with a point calibration source located at a sufficient distance from the detector is that it can neglect coincidence summing effects and get less uncertainty over the calibration process. A more recent example of the ET method with point reference sources is the application of the ETNA code [6]; although, of course, there is no doubt that other researchers also observed those benefits and have applied the ET method using as a reference volume source with a geometry as close as possible to the detector. An extended source was, for example, chosen as the reference source for testing the equivalence of various ET codes in a recently organized inter comparison [7].

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The efficiency of the γ-ray detectors can be calculated using a radioactive source of any geometric shape [radii greater than the detectors faces radii] based on the ET formula. For variety, this one needs to know the geometrical parameters of the source-detector system for the reference and for the unknown sources in order to calculate the effective solid angle of both, as well as the measured efficiency of the detector using a reference source [3].

The main goal of this study is to use the ET formula in order to compute efficiencies for the (NaI & HPGe) detectors by using the uniform volumetric radioactive sources (Marinelli) if a standard source of the same geometry is not available. These formulas were based on reference cylindrical radioactive sources located on the top of the detector surface in order to get the effective solid angle required to apply the method. We then compared these results with the measured one. All of the sources contain aqueous ^{152}Eu radionuclide, which radiates photons with a broad range of energies from 121.78 up to 1408.03 keV. All of the sources have radii greater than the detector's faces radii.

2 Mathematical approach

The effective solid angle of a cylindrical detector used in detecting a photon emitted from an arbitrarily positioned irradiating axial point source was reported [8, 9] as:

$$\Omega_{\mathrm{eff(Point)}}=\int_{\theta}\int_{\varphi} f_{\mathrm{att}}\sin\theta\mathrm{d}\varphi\mathrm{d}\theta, \tag{4}$$

where f_{att} is a factor that determines the photon attenuation by all absorbers between source and detector and is expressed as:

$$f_{\mathrm{att}}=\mathrm{e}^{-\sum_i \mu_i\delta_i}, \tag{5}$$

where μ_i is the attenuation coefficient of the i^{th} absorber for a γ-ray photon with energy E_γ and δ_i is the average γ photon path length through the ith absorber. The effective solid angle for using cylindrical source $\Omega_{\mathrm{eff(Cyl)}}$ can be expressed as [10]:

$$\Omega_{\mathrm{eff(Cyl)}}=\frac{\int_h\int_\alpha\int_\rho S_{\mathrm{f}}.\Omega_{\mathrm{eff(Point)}}\mathrm{d}V}{V} \ \&\mathrm{d}V=\rho\mathrm{d}\rho\mathrm{d}\alpha\mathrm{d}h, \tag{6}$$

where ρ is the lateral distance from the detector axis and makes an angle α with the detector's major axis, h_{o} is the source-detector separation, h is the distance from the point to the end plane of the source, and S_{f} represents the self-attenuation factor of the source contents.

While, the self-attenuation factor S_{f} is given by:

$$S_{\mathrm{f}}=\mathrm{e}^{-\mu_{\mathrm{s}}.d_{\mathrm{s}}}, \tag{7}$$

where μ_{s} is the source attenuation coefficient and d_{s} is the distance traveled by the emitted photon inside the source, as shown in Fig. 1, and d_{s} was found to be a function of the polar and azimuthal angles (θ, φ) inside the source itself [10], and is given by:

$$d_{\mathrm{s_1}}=\frac{h-h_{\mathrm{o}}}{\cos\theta} \text{ for } \theta\leqslant\theta_2' \text{ and } \varphi\leqslant\varphi_{\mathrm{Smax}}', \tag{8}$$

where, the source polar angles θ_1' and θ_2' are given as:

$$\theta_1'=\tan^{-1}\left(\frac{S-\rho}{h-h_{\mathrm{o}}}\right)\&\theta_2'=\tan^{-1}\left(\frac{S+\rho}{h-h_{\mathrm{o}}}\right), \tag{9}$$

where θ_1' and θ_2' are the extreme polar angles of the source and h_{o} is the source-detector separation distance.

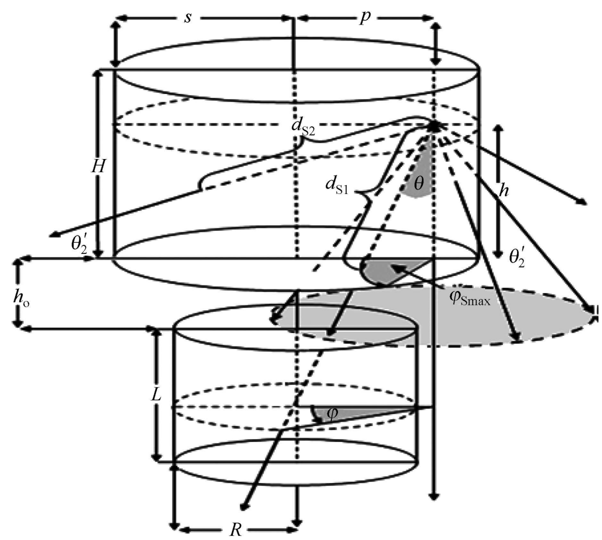

Fig. 1. A cylindrical detector with a cylindrical source of a radius larger than the detector radius.

Where the source azimuthal angle φ_{Smax}' for the photon to escape the source volume is given by:

$$\varphi_{\mathrm{Smax}}'=\cos^{-1}\left(\frac{\rho^2-S^2+(h-h_{\mathrm{o}})^2\tan^2\theta}{2\rho(h-h_{\mathrm{o}})\tan\theta}\right). \tag{10}$$

So, Eq. (6) will be written as:

$$\Omega_{\mathrm{eff(Cyl)}}=\frac{\int_h\int_\alpha\int_\rho f_{\mathrm{att}}.S_{\mathrm{f}}.\Omega_{\mathrm{eff(Point)}}.\rho\mathrm{d}\rho\mathrm{d}\alpha\mathrm{d}h}{V}. \tag{11}$$

Thus, the effective solid angle of a cylindrical detector in the case of a cylindrical source of radius ($S\geqslant R$) and height H can be expressed by:

$$\Omega_{\mathrm{eff(Cyl)}}=\frac{\int_{h_{\mathrm{o}}}^{H+h_{\mathrm{o}}}\int_0^{2\pi}\int_0^{S} f_{\mathrm{att}}.S_{\mathrm{f}}.\Omega_{\mathrm{eff(Point)}}.\rho\mathrm{d}\rho\mathrm{d}\alpha\mathrm{d}h}{\pi S^2 H}. \tag{12}$$

We then rewrite Eq. (12) as follows:

$$\Omega_{\mathrm{eff(Cyl)}}=\frac{1}{\pi S^2 H}\int_{h_{\mathrm{o}}}^{H+h_{\mathrm{o}}}\left(\int_0^{2\pi}\int_0^{R} f_{\mathrm{att}}.S_{\mathrm{f}}.\Omega_{\mathrm{eff(Point_{\rho<R})}}.\rho\mathrm{d}\rho\mathrm{d}\alpha + \int_0^{2\pi}\int_R^{S} f_{\mathrm{att}}.S_{\mathrm{f}}.\Omega_{\mathrm{eff(Point_{\rho\geqslant R})}}.\rho\mathrm{d}\rho\mathrm{d}\alpha\right)\mathrm{d}h. \tag{13}$$

The effective solid angle of the detector $\Omega_{\rm Mar}$ when using a radioactive Marinelli source, according to [11], is:

$$\Omega_{\rm Mar}=\frac{\sum\limits_{i=1}^{5}\Omega_{Vi}.V_i}{\sum\limits_{i=1}^{5}V_i}, \tag{14}$$

where the effective solid angles, as shown in Fig. 2, of each part of the Marinelli beaker Ω_{V1}, Ω_{V2}, Ω_{V3}, Ω_{V4} and Ω_{V5} associated with the volumes V_1, V_2, V_3, V_4 and V_5, respectively, are given by [10]. The volume V_1 acts as a solid cylinder with height L_1 and radius R while the volumes V_2, V_3, V_4 and V_5 act as a thick cylindrical ring with height L_1, h_0, L, and, L_2, respectively, with inner radius S_1 and outer radius S_2.

$$\left.\begin{aligned}
\Omega_1&=\frac{1}{\pi R^2L_1}\int_{h_{\rm o}}^{h_{\rm o}+L_1}\int_0^{2\pi}\int_0^{R}S_{\rm f}.\Omega_{{\rm point}(\rho<R)}\rho{\rm d}\rho{\rm d}\alpha{\rm d}h,\\
\Omega_2&=\frac{1}{\pi(S_2^2-R^2)L_1}\int_{h_{\rm o}}^{h_{\rm o}+L_1}\int_0^{2\pi}\int_R^{S_2}S_{\rm f}.\Omega_{{\rm point}(\rho\geqslant R)}\rho{\rm d}\rho{\rm d}\alpha{\rm d}h,\\
\Omega_3&=\frac{1}{\pi(S_2^2-S_1^2)h_{\rm o}}\int_0^{h_{\rm o}}\int_0^{2\pi}\int_{S_1}^{S_2}S_{\rm f}.\Omega_{{\rm point}(\rho\geqslant R)}\rho{\rm d}\rho{\rm d}\alpha{\rm d}h,\\
\Omega_4&=\frac{1}{\pi(S_2^2-S_1^2)L}\int_0^{L}\int_0^{2\pi}\int_{S_1}^{S_2}S_{\rm f}.\Omega_{{\rm point}(\rho\geqslant R)}\rho{\rm d}\rho{\rm d}\alpha{\rm d}h,\\
\Omega_5&=\frac{1}{\pi(S_2^2-S_1^2)L_2}\int_0^{L_2}\int_0^{2\pi}\int_{S_1}^{S_2}S_{\rm f}.\Omega_{{\rm point}(\rho\geqslant R)}\rho{\rm d}\rho{\rm d}\alpha{\rm d}h.
\end{aligned}\right\} \tag{15}$$

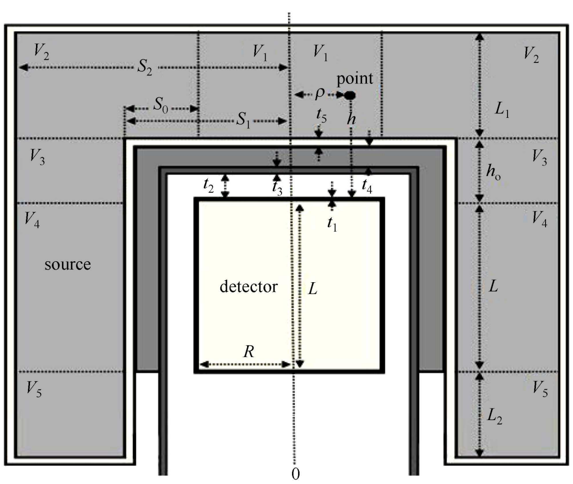

Fig. 2. The geometric configuration of the Marinelli beaker source with (NaI(Tl) & HPGe) detectors.

To determine the absorption of photons through the Marinelli beaker source, there are two factors to be considered. The first is the self-absorption factor of the source medium. For the given Marinelli beaker source and photon energy, the self-absorption is a function of the path length of the photon in the source medium. Table 1 shows that there are two different allowed photon path lengths $d_{\rm s}$ (through the source medium) corresponding to the lateral distance from the detector axis ρ.

The second factor is the attenuation factor of the Marinelli beaker container material, dead layer and end-cap and absorber where the attenuation of the Marinelli beaker container of thickness t_5, the dead layer of thickness $t_{1,}$ the end-cap with thickness t_3, and the absorber of thickness $t_{4,}$ is a function of the photon path length through these materials. Table 1 shows the four different photon path lengths through the Marinelli beaker container, dead layer, end-cap, and absorber material (δt_5, δt_1, δt_3 and δt_4, respectively) corresponding to the lateral distance from the detector axis ρ, while the effect of the air thickness t_2 is neglected.

Table 1. The photon path lengths through the source-detector system.

ρ	$d_{\rm s}$			
$\rho<R$	$(h-h_{\rm o})/\cos\theta$			
$\rho\geqslant R$	$\left[\left(\rho\cos\varphi+\Delta\sqrt{R^2-\rho^2\sin^2\varphi}\right)-S_{\rm o}\right]/\sin\theta$			
ρ	δt_5	δt_1	δt_3	δt_4
$\rho<R$	$\frac{t_5}{\cos\theta}$	$\frac{t_1}{\cos\theta}$	$\frac{t_3}{\cos\theta}$	$\frac{t_4}{\cos\theta}$
$\rho\geqslant R$	$\frac{t_5}{\sin\theta}$	$\frac{t_1}{\sin\theta}$	$\frac{t_3}{\sin\theta}$	$\frac{t_4}{\sin\theta}$

Now, the full energy peak efficiency (FEPE) of the γ-detectors using a converse well radioactive source can be calculated according to the equations (2, 13 and 14) and based on the reference efficiency of the detectors with respect to reference cylindrical radioactive sources located on the top of the detector surfaces, as follows :

$$\varepsilon_{\rm Mar}(E_\gamma)=\frac{\Omega_{\rm Mar}(E_\gamma)}{\Omega_{\rm eff(Cyl)}(E_\gamma)}\varepsilon_{\rm ref}(E_\gamma), \tag{16}$$

where $\varepsilon_{\rm Mar}(E_\gamma)$ and $\varepsilon_{\rm ref}(E_\gamma)$ are the (FEPE) for the γ-ray detectors using a converse well, a radioactive source, radioactive sources, and a cylindrical radioactive source as a reference geometry, respectively, while $\Omega_{\rm Mar}(E_\gamma)$ and $\Omega_{\rm eff(Cyl)}(E_\gamma)$ are the effective solid angles subtended by the detector surface with both source geometries, respectively [8]. All the integrals encountered are elliptic integrals and do not have a closed form solution, so a numerical solution is obtained using the trapezoidal rule. Although the accuracy of the integration increases with an increase in the number of intervals, the integration converges well, as n=30. A computer program (which was produced using Microsoft's Basic programming language) has been written to calculate the effective solid angles for arbitrary located volumetric sources based on the derived equations.

3 Experimental technique

The full energy peak efficiency (FEPE) for NaI(Tl) scintillation and HPGe detectors were measured at Prof. Dr. Younis. S. Selim's Laboratory for Radiation Physics, Department of Physics, Faculty of Science, Alexandria University. Both detectors are nearly of the same volume, and have geometry parameters provided by manufacturers listed in Table 2. Both detectors were calibrated by using converse well radioactive sources and cylindrical radioactive sources as reference geometry.

The values of half-life, photon energies and photon emission probabilities per decay for the radionuclide used in the calibration process, which are available from the National Nuclear Data Center Web Page or on the IAEA website, plus the volumetric sources activities and their uncertainties for all sources used, are listed in Table 3. Finally, the dimensions of the used cylindrical containers and Marinelli beakers are given in Table 4.

Table 2. Set up parameters with acquisition electronics specifications for NaI(Tl) and HPGe detectors.

items	HPGe [Det1]	NaI(Tl) [Det2]
manufacturer	Canberra	Canberra
serial number	06089367	09L 654
detector model	GC1520	802
type	closed end coaxial	cylindrical
mounting	vertical	vertical
resolution (FWHM)/keV	2 at 133	7.5% at 661
cathode to anode voltage	+4500V DC	+1100 V DC
shaping mode	Gaussian	Gaussian
crystal diameter/cm	4.8	5.08
crystal length/cm	5.45	5.08
top cover thickness/cm	Al (0.05)	Al (0.05)
side cover thickness/cm	Al (0.05)	Al (0.05)
reflector – oxide/cm	—	0.25

Table 3. Half lives, photon energies and photon emission probabilities per decay for the radionuclides used in this work plus the activities of the used uniform sources and their uncertainties.

the half life, photon energies and photon emission probabilities per decay			
PTB nuclide	energy/keV	emission probability (%)	half life/day
	121.78	28.4	
	244.69	7.49	
	344.28	26.6	
^{152}Eu	443.97	2.78	4943.29
	778.9	12.96	
	964.13	14.0	
	1408.03	20.87	
volumetric sources activities and their uncertainties			
sources	activity/kBq±1.98%	reference date	purchasing company
$V1$			Nalgene
$V2$			Nalgene
$V3$	5	1 Jan 2010	Nalgene
$M1$			Ga-Ma & Associates, Inc
$M2$			Ga-Ma & Associates, Inc
$M3$	10		nuclear technology services, Inc

Table 4. Dimensions of the uniform radioactive cylindrical and Marinelli beaker sources in cm.

cylindrical sources			
volume/cm^3)	outer diameter	height	wall thickness
$V1$ (200)		2.1	
$V2$ (300)	11.23	3.2	0.20
$V3$ (400)		4.2	

marinelli beaker sources					
volume/cm^3	outer diameter	height of source	wall thickness	height of well	well diameter
$M1$ (500)	11.7	7.9	0.15	6.8	7.7
$M2$ (200)	11.4	4.9	0.26	3.8	7.8
$M3$ (1000)	14.0	8.3	0.10	6.1	8.5

4 Results and discussion

The measured efficiency values as a function of the photon energy $\varepsilon(E)$, for both NaI(Tl) and HPGe detectors were calculated using the following formula:

$$\varepsilon(E)=\frac{N(E)}{T\cdot A_{\rm S}\cdot P(E)}\prod C_{\rm i}, \tag{17}$$

where $N(E)$ is the number of counts in the full-energy peak, which was obtained using Genie 2000 software, T is the measuring time (in seconds), $P(E)$ is the photon emission probability at energy E, which was obtained from Genie 2000 standard library, while, $A_{\rm S}$ is the radionuclide activity and $C_{\rm i}$ represents the correction factors due to dead time and radionuclide decay.

In order to minimize the effect of the dead time and the pile up effects, the activity of the sources was low enough (see Table 3) to avoid high count rates when measuring at low distance, which implies that there is a long counting time at high distance.

No summing correction was done due to the very low dead time associated with measurements. The corresponding correction factor for the dead time was obtained simply using (ADC) live time. However, the background subtraction was done which was extremely important for low activity sources. The decay correction $C_{\rm d}$ for the calibration source from the reference time to the run time was given by:

$$C_{\rm d}=\exp(\lambda\cdot\Delta T), \tag{18}$$

where λ is the decay constant and ΔT is the time interval over which the source allowed to decay until the run time. The main source of uncertainty in the efficiency calculations were the uncertainties of the activities of the standard source solutions. The uncertainty in the (FEPE) σ_ε was considered and is given by:

$$\sigma_\varepsilon=\varepsilon.\sqrt{\left(\frac{\sigma_N}{\partial N}\right)^2+\left(\frac{\sigma_A}{\partial A}\right)^2+\left(\frac{\sigma_P}{\partial P}\right)^2}, \tag{19}$$

where σ_N, σ_A and σ_P are uncertainties associated with the uncertainties in the quantities $N(E)$, $A_{\rm S}$ and $P(E)$, respectively.

The percentage deviations between the calculated (with and without self-absorption) and the measured full energy peak efficiency values are calculated by:

$$\Delta\%=\frac{\varepsilon_{\rm cal}-\varepsilon_{\rm meas}}{\varepsilon_{\rm cal}}\times 100, \tag{20}$$

where $\varepsilon_{\rm cal}$ and $\varepsilon_{\rm meas}$ are the calculated and experimentally measured efficiencies, respectively.

The relation between the calculated (FEPE) due to transfer from ($V1$, $V2$ and $V3$) to ($M1$, $M2$ and $M3$) based on Eq. (16) and the measured ones based on Eq. (17), respectively, for the (NaI & HPGe) detectors with their associated uncertainties as a function of the photon energy using a reference cylindrical radioactive sources ($V1$, $V2$ and $V3$) and Marinelli radioactive sources ($M1$, $M2$ and $M3$) are depicted in Figs. 3 to 8. The figures show that the efficiency of both (NaI & HPGe) detectors using Marinelli beaker as a container of radioactive material is higher than that obtained by the other volumetric cylindrical sources.

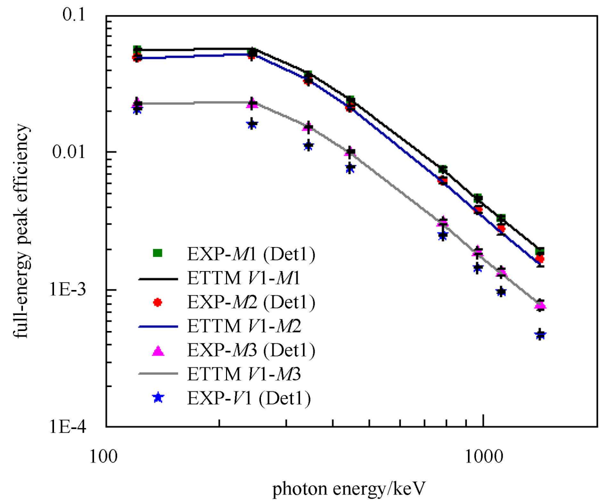

Fig. 3. The measured full-energy efficiency values with their associated uncertainties as a function of the photon energy for HPGe detector using ($V1$, $M1$, $M2$ and $M3$) and the calculated ones due to transfer from $V1$ to $M1$, $M2$ and $M3$.

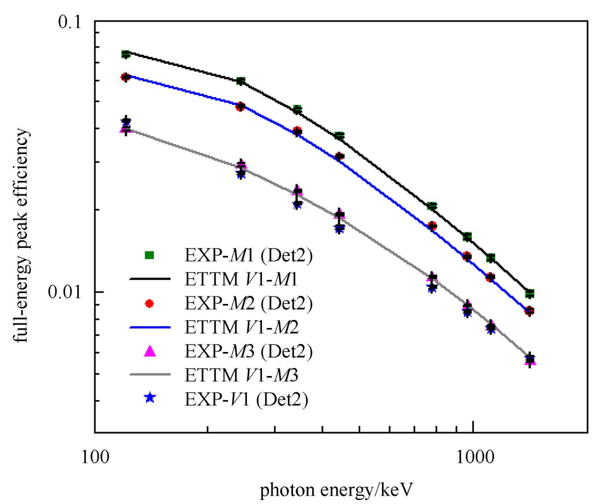

Fig. 4. The measured full-energy efficiency values with their associated uncertainties as a function of the photon energy for NaI detector using ($V1$, $M1$, $M2$ and $M3$) and the calculated ones due to transfer from $V1$ to $M1$, $M2$ and $M3$.

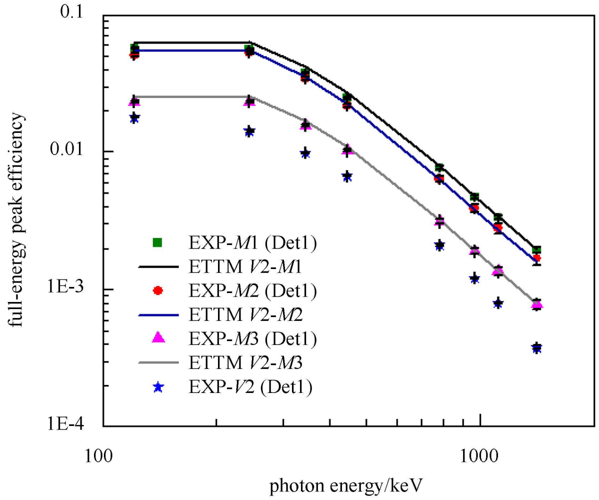

Fig. 5. The measured full-energy efficiency values with their associated uncertainties as a function of the photon energy for HPGe detector using ($V2$, $M1$, $M2$ and $M3$) and the calculated ones due to transfer from $V2$ to $M1$, $M2$ and $M3$.

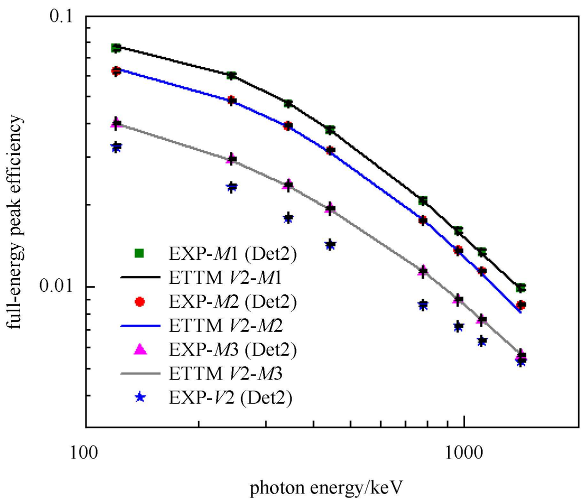

Fig. 6. The measured full-energy efficiency values with their associated uncertainties as a function of the photon energy for NaI detector using ($V2$, $M1$, $M2$ and $M3$) and the calculated ones due to transfer from $V2$ to $M1$, $M2$ and $M3$.

There is a relative difference between the measured and the calculated efficiency values, which reaches to 7% as a maximum value, that indicates the success of the ET methodology in general at very close distances and for higher energies. This is due to the large detector's length, which gives it a reasonable efficiency for the highest energy gamma-rays, (the contribution to the full-energy peak from the Compton process is large with large detectors crystal and at lower distances from the detector surface), the change in solid angle (the efficiency decreases with increasing the source volume), the interaction of gamma-ray with the detector's material plus its type (HPGe has a lower atomic number than NaI(Tl). For this reason, the HPGe detector has smaller interaction probabilities for photons and, therefore, smaller relative efficiency) and both top and side of the detector is a more sensitive region than its side or top separately, where the sources in these case fitted well closer to the detector as the Marinelli beaker. In addition, the relative difference is due to the accuracy problem in the measuring technique using the volumetric sources with the detectors, which depends on the fine-tune adjustment problem with the detector's parameters and the geometry of the instrument used in the ET formula.

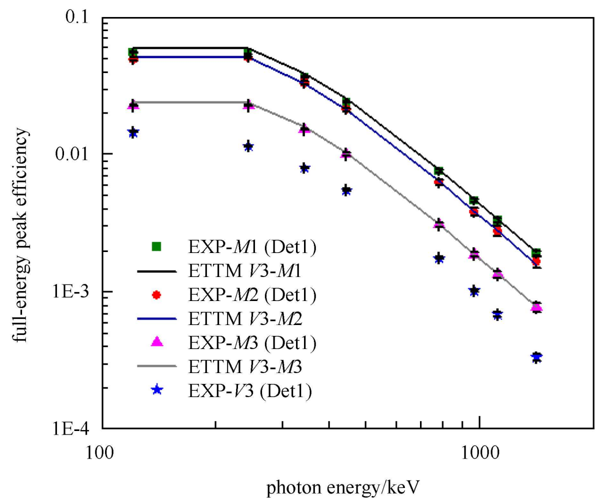

Fig. 7. The measured full-energy efficiency values with their associated uncertainties as a function of the photon energy for HPGe detector using ($V3$, $M1$, $M2$ and $M3$) and the calculated ones due to transfer from $V3$ to $M1$, $M2$ and $M3$.

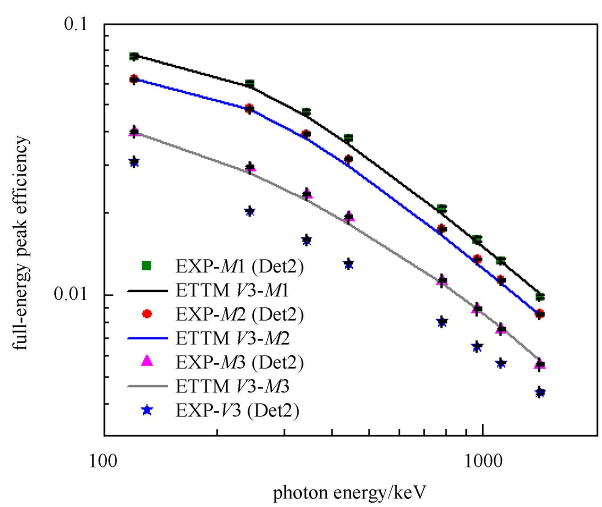

Fig. 8. The measured full-energy efficiency values with their associated uncertainties as a function of the photon energy for NaI detector using ($V3$, $M1$, $M2$ and $M3$) and the calculated ones due to transfer from $V3$ to $M1$, $M2$ and $M3$.

The present work provides a greater understanding of several aspects of gamma-ray spectroscopy and provides a useful tool (ET formula) for efficiency computation for γ-ray detectors. This tool constitutes a good approach for the efficient computation for laboratory routine measurements and can save time by avoiding experimental calibration for different sample geometries, where the method takes into account the enable to improve both efficiency calibration and activity measurement accuracy. This method is fully sufficient for most routine measurement work under one condition: the geometrical parameters of the source-detector system must be provided exactly.

5 Conclusion

This work leads to a simple ET formula to evaluate the full-energy peak over a wide energy range, which deals with (NaI(Tl) & HPGe) detectors for using a converse well radioactive sources (Marinelli beaker), the passage length traveled by a photon within the active medium of the source, as well as the geometrical solid angle subtended by the source to the detector and the attenuation of photons by the source itself. The source container, the detector end-cap and holder material are also presented by a simple formula. A good agreement between the measured and calculated efficiencies for the γ-ray detectors were observed from the data comparisons. Therefore, the present approach shows a considerable possibility for calibrating the detectors through the determination of a full energy peak efficiency curve, even in those cases when no standard source is available, which is considered as the final goal of this work.

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