

## A METHOD OF EFFICIENCY CALIBRATION FOR DISK SOURCES IN GAMMA-RAY SPECTROMETRY

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*Abstract.* The quality of the results of gamma spectrometry measurement depends directly on the accuracy of the detection efficiency in the specific measurement conditions. Experimental efficiency calibration is restricted to several measurement geometries and can not be applied directly to all measurement configurations. In this work an approach using efficiencies measured with point sources combined with theoretical procedures is applied for obtaining the peak efficiency  $\varepsilon(E)$  for disk sources measured with a NaI(Tl) detector. Coincidence summing effects are evaluated using GESPECOR and are used to correct the experimental values of the efficiencies. The resulting efficiencies were compared with predictions of the ETNA (Efficiency Transfer for Nuclide Activity measurements) software. The efficiency curves obtained in this way will be applied to the measurement of samples from the decommissioning of the VVR-S reactor.

*Key words:* gamma-ray spectrometry, detector characterization, efficiency calibration.

### 1. INTRODUCTION

An important step in the implementation of the decommissioning plan of the VVR-S nuclear reactor belonging to the National Institute for Physics and Nuclear Engineering “Horia-Hulubei” (NIPNE H-H), Bucharest, Romania, is represented by the radiological characterization of the reactor. The Radiological Characterization Laboratory from NIPNE H-H has the principal task to achieve the radiological characterization in different phases of the decommissioning process. This task is very important because it provides the basis for the correct classification of various types of wastes, which in turn affects the decommissioning solution and the associated costs. The measurement method

should be reliable and efficient. Furthermore, it should be flexible, able to provide accurate results for the variety of samples, with different compositions and densities, with different shapes and possibly non-uniform activity distribution that should be assessed. The appropriate efficiency calibration in the above conditions is a challenging task.

In this work an approach using the experimental efficiency measured with point sources combined with theoretical procedures is applied for obtaining the peak efficiency  $\varepsilon(E)$  for disk sources measured with a NaI(Tl) detector. NaI(Tl) detectors are commonly used to identify and measure activities of low-level radioactive sources. They have high detection efficiency and operate at room temperature [1]. One of the most important parameters in the calculation of the gamma activity is the detection efficiency which is usually determined by using calibrated standard sources. This technique is very convenient for the case of point sources measured far from the detector. However, in the case of extended sources, measured close to the detector, it is difficult to find appropriate calibration sources; the coincidence summing effects, expected to be high in close to detector measurement configurations, additionally complicate the problem.

## 2. EXPERIMENTAL MEASUREMENTS

The gamma spectrometry system used consisted of a ScintiPack Photomultiplier Base with Preamplifier and High Voltage Supply type 296 and a Digital Portable Multichannel Analyzer type DigiDART. The detector is a 3“X3” NaI(Tl) scintillator, with an energy resolution of 70.62 keV at 1332 keV ( $^{60}\text{Co}$ ). The recommended operating bias is + 1000V.

First, the detector efficiencies were determined experimentally, as a function of gamma-ray energies [2], using  $^{241}\text{Am}$ ,  $^{152}\text{Eu}$ ,  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  point sources. The reference activities and the uncertainties (1T) of the used point sources were of (2029±20) Bq for  $^{241}\text{Am}$ , (6305±63) Bq for  $^{152}\text{Eu}$ , (3805±38) Bq for  $^{137}\text{Cs}$  and (8235±82) Bq for  $^{60}\text{Co}$ .

The sources were placed at 8 mm from the face of the NaI(Tl) detector. The measurements were performed in the horizontal plane, parallel with the entrance face of the detector, at radial distances  $r = 0, 1, 2, 3$  and 4 cm from the detector axis. The experimental arrangement is depicted in Fig. 1.

For these measurements the detector was collimated. A set of data was collected for this measurement geometry. The experimental values of the efficiencies are represented in Fig. 2. On this figure it is seen that the experimental efficiencies  $\varepsilon(E)$  do not present a smooth variation with the energy  $E$ .

Fig. 1 – Experimental set-up, with the point sources at distances  $r = 0, 1, 2, 3, 4$  cm from the detector axis, in a horizontal plane at 8 mm from the detector face.

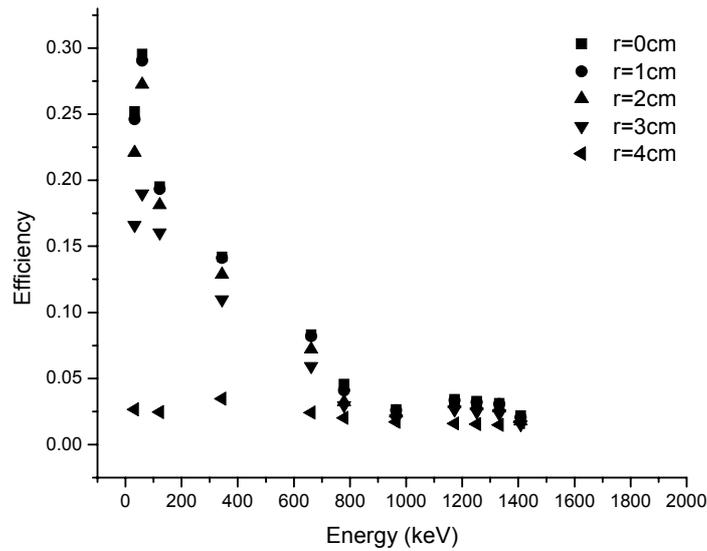
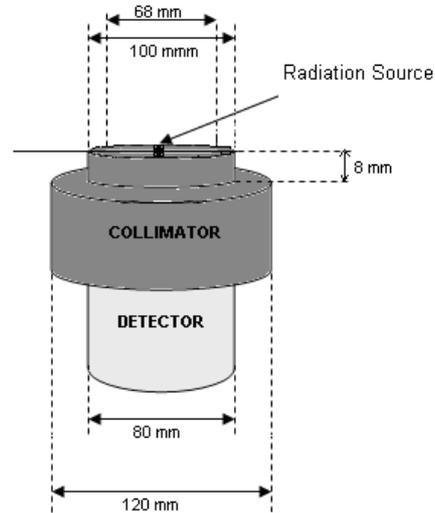


Fig. 2 – The experimental values of the detection efficiency for point sources.

### 3. COINCIDENCE SUMMING CORRECTIONS

The origin of the deviation of the efficiency data displayed in Fig. 2 from smooth curves as a function of energy is the presence of important coincidence summing effects in the case of  $^{60}\text{Co}$  and  $^{152}\text{Eu}$  sources. Because these effects are

peak and nuclide specific, it is necessary to take them into account in order to obtain a generally useful efficiency curve. A realistic evaluation of the coincidence summing effects is a difficult task, especially in the case of nuclides with complex decay schemes like  $^{152}\text{Eu}$ . It implies an intricate combination of decay scheme parameters with peak and total efficiencies specific to the measurement conditions. The best method available for this purpose is the Monte Carlo method. In this work in order to evaluate the coincidence summing corrections we applied a dedicated software called GESPECOR.

GESPECOR [3] is a Monte Carlo simulation code specifically developed for the computation of efficiency, of matrix effects [4] and of coincidence summing effects [5] in gamma-ray spectrometry with HPGe detectors. The GESPECOR software is a realistic simulation program that can describe in detail the physics processes and the measurement arrangement; it also incorporates efficient algorithms and variance reduction techniques and furthermore it has a user friendly interface.

As it is devoted to germanium detectors, GESPECOR cannot be directly applied in the case of a NaI(Tl) detector. In the present work we have evaluated the coincidence summing correction factors for the NaI(Tl) detector by combining the decay scheme data evaluated by GESPECOR with experimental values of the peak and of the total efficiencies for the point source measurements, in an iterative procedure. The procedure is briefly presented below.

Schematically, the coincidence summing effects result in two types of changes in the count-rate from a given peak. Consider a peak of energy  $E$ . In the case when a photon of energy  $E$  delivers its complete energy in the detector a pulse in the peak is always registered in the absence of coincidence summing effects. However, if another photon, with energy  $E_1$ , interacts simultaneously with the detector, then the pulse is lost from the peak, being moved to a higher energy channel, corresponding to the total energy delivered in the detector by both photons. The probability of such *coincidence losses* from the peak of energy  $E$  depends on the probability of other photons being emitted simultaneously with the photon with energy  $E$  (that is, depends on the decay scheme of the nuclide) and on the total efficiency of the detector for the energy of the other photons. In the case when the transition resulting in the emission of the photon with energy  $E$  can also take place in successive transitions in which photons with energies  $E_1$  and  $E_2$ ,  $E_1+E_2=E$ , are emitted, then simultaneous total energy deposition of the two photons in the detector would add a pulse in the peak of energy  $E$ . The probability of such *summing in* (sum peak) effects depends on the probability of emission of the corresponding group of photons and on the peak efficiency for the energies of these photons.

According to the above discussion, in order to correct the efficiency values for the peaks of  $^{152}\text{Eu}$ , the peak and the total efficiency for the energies of other

photons emitted by  $^{152}\text{Eu}$  is required. For example, in the case of the peak with energy  $E=121.78$  keV, only coincidence losses are possible. The losses are produced when any photon from a list containing 71 photons which can be emitted simultaneously with the photon of energy 121.78 keV interacts with the detector. These photons span an energy range from the X ray region to 1647 keV and consequently the total efficiency for the energy in this range is required. In the case of the peak with energy 1408.01 keV there are 21 combinations of various photons which can contribute to sum peak effects; the peak efficiency for all these photons is required for calculating the coincidence summing corrections for the 1408.01 keV peak.

As the correct peak and total efficiency values required for performing coincidence summing calculations are not known in advance, we have applied an iterative procedure.

In the first iteration the directly measured values of the peak efficiencies were used for the computation of summing in corrections. The total efficiencies could be measured directly only for  $E=59.54$  keV ( $^{241}\text{Am}$ ), 661.66 keV ( $^{137}\text{Cs}$ ) and 1251.87 keV (the mean energy of the closely enough lines of  $^{60}\text{Co}$ ). Using the fact that the ratio between the peak efficiency and the total efficiency is a smooth function of energy, a first estimate of the total efficiency as a function of energy could be obtained (Fig. 3) even if only few directly measured total efficiency data were available.

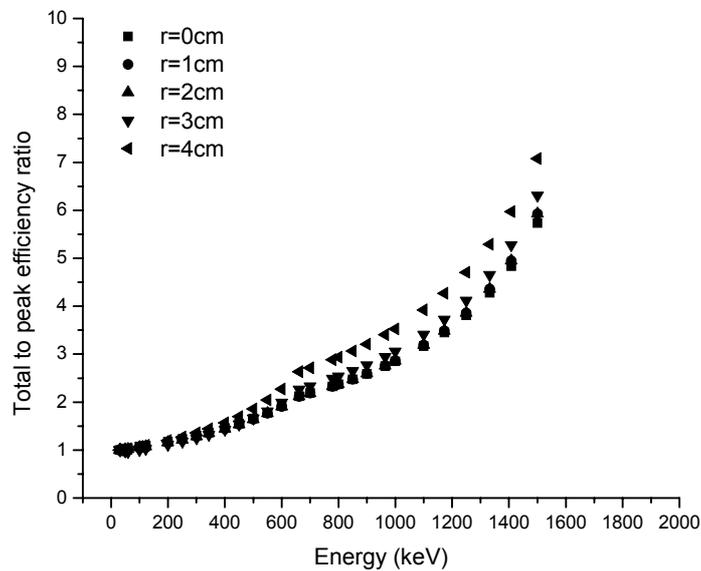


Fig. 3 – Experimental ratio of total efficiency to peak efficiency *versus* the energy.

The values of the peak and total efficiencies obtained in this way were used for calculating the coincidence summing correction factors in the first iteration. These correction factors were subsequently used to obtain improved values of the peak efficiencies (second iteration). The total efficiency curve in the second iteration was obtained using the second iteration values of the peak efficiency and the measured values of the total efficiency in the same way as in the case of first iteration (Fig. 4). The values of the peak and total efficiency obtained in the second iteration were used to compute the coincidence summing correction factors in the second iteration. It was not necessary to do the computations in higher order iterations and the final values of the peak efficiencies were obtained from the measured values of the peak efficiencies and the coincidence summing correction factors in the second iteration.

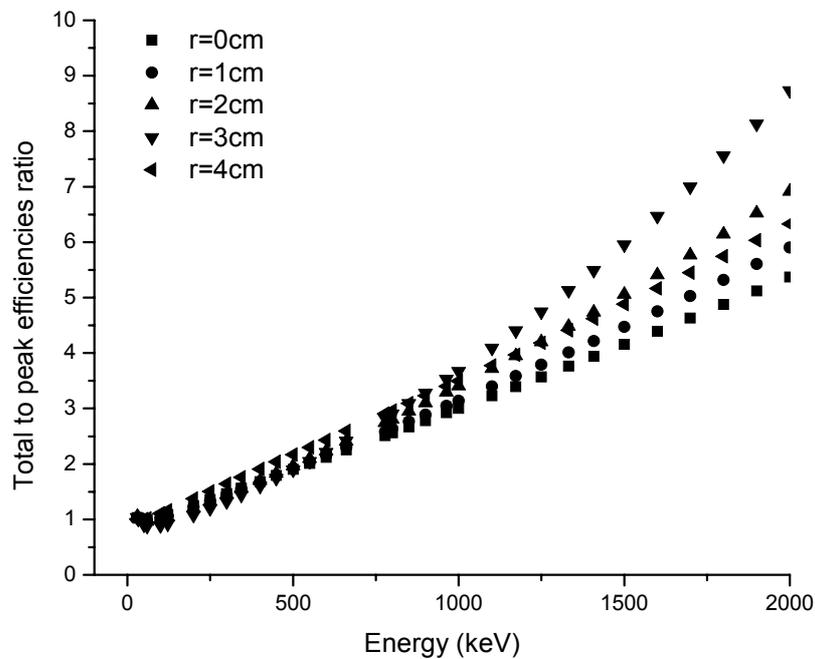


Fig. 4 – Ratio of total efficiency to peak efficiency *versus* the energy after applying the coincidence summing corrections in the first iteration.

The finally adopted values of the peak efficiencies for the point source measurements are represented in Fig. 5.

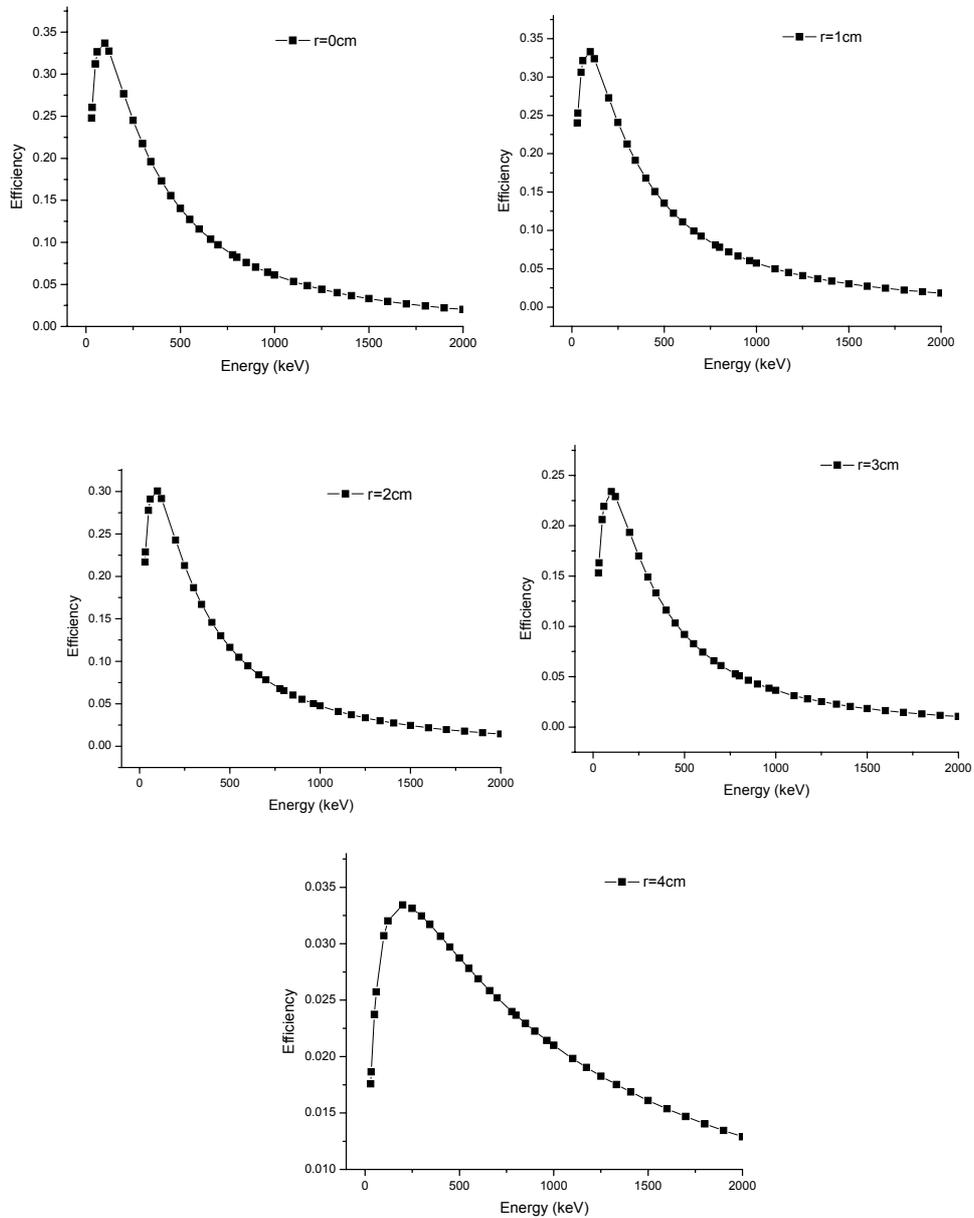


Fig. 5 – The detection efficiency for point sources measured in horizontal plane at 8 mm from the face of the detector, corrected for the effects of coincidence summing.

In Table 1 the coincidence summing correction factors computed in the first and second iteration for the five measurement geometries are presented.

*Table 1*

The coincidence summing correction factors					
Energy (keV)	r = 0cm	r = 1cm	r = 2cm	r = 3cm	r = 4cm
First iteration					
121.78	0.694	0.702	0.728	0.784	0.905
344.28	0.906	0.912	0.924	0.931	0.955
778.90	0.826	0.831	0.852	0.875	0.954
964.13	0.641	0.649	0.679	0.749	0.959
1173.24	0.866	0.867	0.876	0.892	0.921
1332.50	0.882	0.884	0.890	0.902	0.932
1408.01	0.670	0.678	0.707	0.773	0.970
Second iteration					
121.78	0.659	0.666	0.692	0.757	0.909
344.28	0.872	0.874	0.885	0.903	0.951
778.90	0.772	0.779	0.802	0.849	0.951
964.13	0.601	0.611	0.644	0.739	0.955
1173.24	0.884	0.884	0.893	0.902	0.926
1332.50	0.873	0.874	0.883	0.895	0.928
1408.01	0.635	0.646	0.677	0.768	0.966

The point source efficiencies obtained in this way can be used to evaluate the efficiency for other types of sources. In the next section the case of disk sources will be presented.

#### 4. COMPUTATION OF THE EFFICIENCY FOR DISK SOURCES

In the radiological characterization of the VVR-S reactor frequently disk sources are evaluated by gamma spectrometry for surface contamination. Due to the lack of appropriate certified disk sources to be used for efficiency calibration, we computed the efficiency for disk sources on the basis of point source efficiencies.

Assuming cylindrical symmetry of the detector, the efficiency for a disk source of radius  $R$  can be computed using the formula:

$$\varepsilon = \frac{2\pi \int_0^R \varepsilon_r r dr}{\pi R^2} \quad (1)$$

where  $\varepsilon_r$  is the efficiency for a point source located in the plane of the disk source at the distance  $r$  from the axis of the detector. The computed efficiency curve for a disk source of radius  $R = 4$  cm obtained using the  $\varepsilon_r$  values from Fig. 5 is represented in Fig. 6.

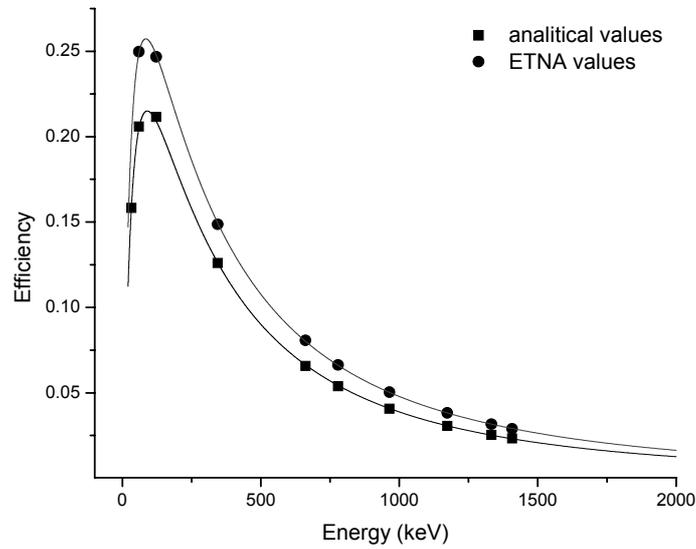


Fig. 6 – Comparison between the efficiency values obtained by Eq. (1) and by ETNA in the case of a disk source with radius  $R = 4$  cm.

We have also estimated the efficiency for the disk source using the efficiency transfer method. In this method the efficiency for a given measurement is computed by the product of the transfer factor and the measured efficiency for a reference measurement. The transfer factor can be computed by Monte Carlo methods [6] or by simplified procedures [7, 8] as the ratio between the computed efficiency for the given measurement and the efficiency for the reference measurement. The advantage of this procedure is the fact that the results are only marginally affected by the uncertainty of the detector data, which is a major source of uncertainty in the direct computation of efficiency. Indeed, an incorrect value e.g. of the detector radius will affect in the same way the computed efficiency both for the measurement of interest and for the reference measurement; being a ratio of the two efficiencies, the transfer factor is much less sensitive to the fact that a

wrong value was used in the computation. Clearly the method of efficiency transfer gives best results in the case when the measurement of interest is similar with the reference measurement.

The National Laboratory Henri Becquerel (LNHB), Saclay, France, developed a few years ago the Efficiency Transfer for Nuclide Activity (ETNA) software [7, 8] for calculating the detector efficiency under measurement conditions different from those of calibration, and for correcting coincidence-summing effects.

Using this software, the detector efficiency for disk sources was calculated for the same detector-source geometry. The reference measurement is represented by the point source measurement with the source located at  $r = 0$  cm from the axis of the detector; the reference efficiency is then  $\varepsilon_r$ , for  $r = 0$  cm.

In Fig. 6, a comparison between the results obtained using formula (1) and ETNA values of the efficiency in case of the disk source is presented.

The analytical values of the efficiency and the ETNA prediction are in reasonable agreement. The differences between them derive from the input parameters in the two cases and the different method used. In the computations carried out with ETNA the information on the efficiency for other radial distances, besides  $r = 0$  cm, cannot be included.

## 5. CONCLUSIONS

The quality of gamma spectrometry measurements depends directly on the knowledge of the detection efficiency and the geometrical conditions of the source-detector arrangement. In this work an approach using the experimental efficiency measured with point sources combined with theoretical procedures was applied for obtaining the peak efficiency  $\varepsilon(E)$  for disk sources measured with a NaI(Tl) detector. The coincidence summing effects were evaluated using GESPECOR software and were used to correct the experimental values of the efficiencies. Using these results an analytical procedure was implemented to calculate the efficiency for disk sources. The results of disk efficiencies were compared with predictions of the ETNA software. The efficiency curve obtained in this way will be applied to the measurement of samples from the decommissioning of the VVR-S reactor.

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