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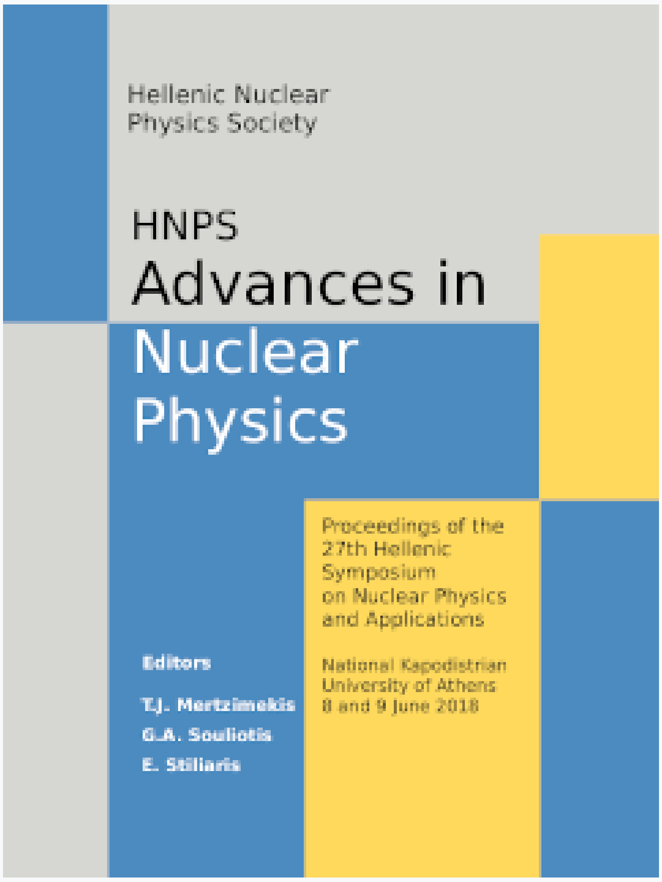

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Development of techniques based on Monte Carlo simulations for clearance of Co-57 and Ge-68/Ga-68 sealed radioactive sources

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Abstract Sealed radioactive sources of Co-57 and Ge-68/Ga-68 are used for the calibration of various nuclear medicine systems like Gamma camera and PET imaging. After their useful life, these sealed sources need to be kept in control for decay until meeting the general clearance criterion. The aim of this work is to determine the activity of spent sealed radioactive sources of Ge-68 / Ga-68 and Co-57. For this purpose, techniques based on Monte Carlo simulation by the use of the MCNPX code was developed for evaluation of the 3”x3” NaI(Tl) scintillator efficiency for specific source–detector geometries. These techniques proved to be accurate.

Keywords Sealed sources, Clearance, NaI(Tl), Gamma-ray spectrometry, MCNPX simulations

INTRODUCTION

Nowadays there is a variety of sealed radioactive sources, such as Co-57 and Ge-68/Ga-68, which are used for the precise calibration of nuclear medicine systems. After their useful life, these sources need to be handled and kept into storage until they meet the general clearance criterion. In some cases, the sources certificates with their characteristics and nominal activity have been lost, so their current activity cannot be estimated. For the experimental determination of the source activity, the detector efficiency should be evaluated for the specific source-detector geometry. In case of complex geometries, a common approach to perform the efficiency calibration is Monte Carlo simulation techniques. Thus, in the specific study, a semi-empirical method was developed based on Monte Carlo simulation by utilizing the MCNPX code for evaluation of the 3”x3” NaI(Tl) scintillator efficiency for specific source-detector geometries.

For this purpose, gamma-ray spectrums were taken for different acquisition time by a NaI(Tl) detector for two types of sealed radioactive sources: 1) a flood source containing Co-57; 2) a line source containing Ge-68/Ga-68. For these source-detector geometries and the specific gamma ray energies, simulations were performed with the MCNPX code. The MCNPX models were validated by the certified activities of radiation sources of these types.

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MATERIALS AND METHODS

Gamma-ray spectrums were taken for different acquisition time by the 3'x3' NaI(Tl) detector for two types of sealed radioactive sources:

i. a line source containing Ge-68/Ga-68 (Fig.1, left)
ii. a flood source containing Co-57 (Fig.1, right)

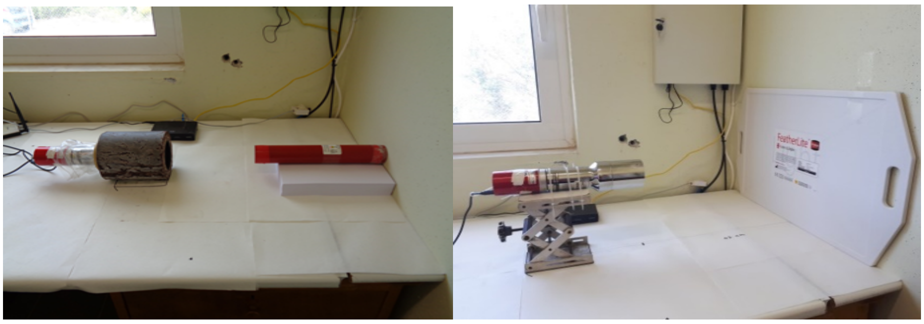

Fig. 1. Experimental setup for Ge-68/Ga-68 line source (left) and Co-57 flood source (right)

The gamma spectrometry system is consisted of the following main parts:

- Detector unit: A Bicron Monoline scintillation detector NaI(Tl) (Model 3M3/3) with a 3'x3' crystal in a thick aluminum housing, covered with a white reflector, including a photomultiplier tube, an internal magnetic/light shield and a 14-pin connector
- Electronics and acquisition unit: a digital signal processing unit (Osprey Digital Tube Base MCA) and a high voltage supply system (850 V)
- Genie^{TM} 2000 spectroscopy software for spectrum acquisition [1]

Gamma-ray spectrums of the two sources were taken for several acquisition times, 7 hours, 1 hour and 15 minutes for the Ge-68/Ga-68 line source and for 7 hours, 2 hours and 15 minutes for the Co-57 flood source. Ge-68 decays by electron capture to Ga-68 which is mainly a positron emitter (89.1%) leading to Zn-68. Then, the excited state of the daughter nucleus Zn-68 decays by a gamma emission of 1077 keV energy (2.93%) [2] (Fig. 2). Co-57 decays by electron capture to the excited state of Fe-57 and subsequently γ photons with energies 122 keV (85.6%), 136 keV (10.68%) and 692.03 keV (0.157%) are emitted leading to the ground state of Fe-57 [2]. The characteristic gamma ray peaks in the resulted spectrums were analyzed with the software package SPECTRW [3]. For the Co-57 source, deconvolution of the peak energies 122 keV and 136 keV was carried out by using this software package [3] (Fig. 2).

For the source-detector geometries and the specific gamma ray energies, simulations were performed by the MCNPX code for evaluation of the NaI(Tl) detector efficiencies. The detector main axis of symmetry was placed perpendicular to the sources flat surfaces. The sources were placed at such distances from the detector, in order acceptable dead time (less than 5%) to be achieved (Fig. 3).

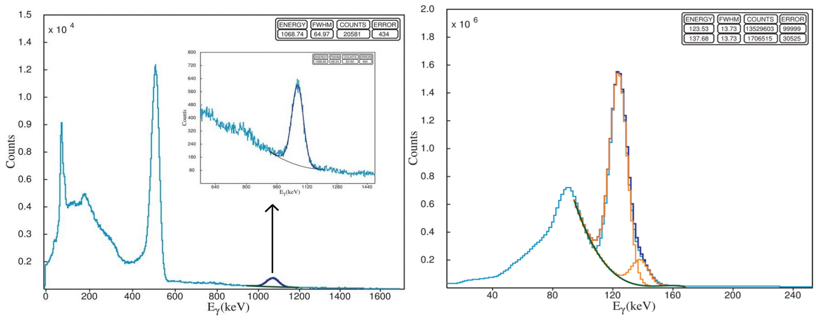

Fig. 2. Ge-68/Ga-68 line source spectrum, 1h exposure (left) and Co-57 flood source spectrum, 2h exposure (right)

Specifically, the Ge-68/Ga-68 line source (ceramic, height 16.3cm, radius 0.07cm) was measured with the cylindrical Pb shielding (height 37cm, radius 2.85cm, thickness 1.52cm) for radiation protection purposes. Pb cylindrical shielding was placed around the detector to reduce the background radiation (height 18cm, radius 7.5cm, thickness 3.3cm). The shielding of the source was placed at the distance of 25cm from the detector (Fig. 1 & 3). The Co-57 flood source (length 61cm and height 41.9cm) was placed at the distance of 47cm from the detector (Fig. 1 & 3).

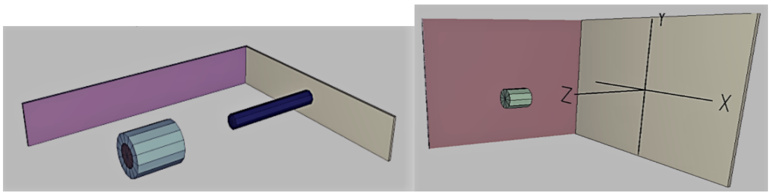

Fig. 3. Ge-68/Ga-68 line source MCNPX visualization (left) and Co-57 flood source MCNPX visualization (right)

Calculations for the nominal activity of the sources based on the certificates information were performed using the law of radioactive decay: $I = I_o \cdot e^{-\lambda t}$, where $\lambda = \frac{ln2}{T_{1/2}}$, I_o = reference radioactivity, t = the time passed from the reference date. The experimental activity was calculated using the formula: $A = \frac{Counts}{(efficiency)\cdot(acquisition\ time)\cdot(fraction\ yield)}$.

RESULTS AND DISCUSSION

The results of the study show that the acquisition time of two hours provided adequate statistics for the activity determination of the flood source Co-57. The determined activity has a deviation of 13,6% from the source nominal activity. Also, one hour measurement was adequate for the activity determination of the line source Ge-68, with a deviation of 26,9% from the nominal activity (Fig. 2 & 4).

The deviation between the evaluated and nominal activities can be attributed mainly to the 15% uncertainty of the certified sources activities. Considering the half-life of Co-57 and Ge-68, which is 0.74 y, this deviation is acceptable. After meeting the general clearance criterion, the sources can be kept in the interim storage additionally for few more months before release from regulatory control.

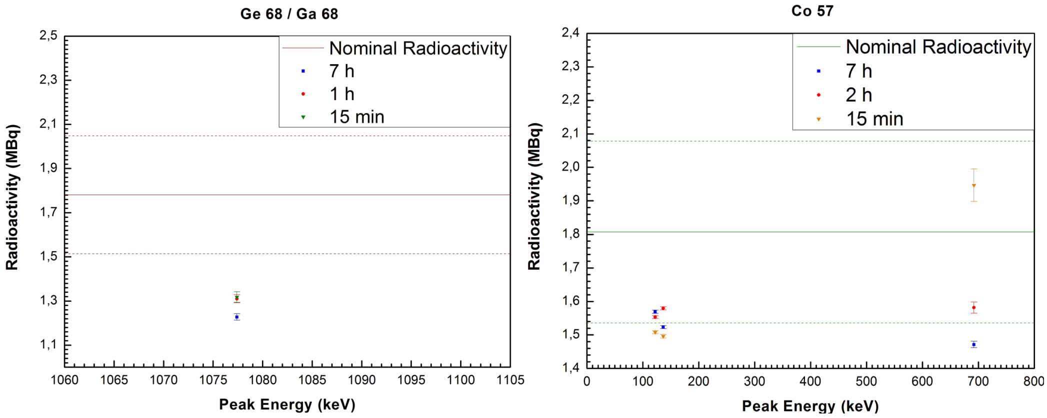

Fig. 4. Determined activities for different acquisition time compared with the nominal activity

Furthermore, the minimum detectable activities (MDA) and the specific experimental set-up are given in Table 1 [4]. The general clearance criterion for Co-57 is 1 Bq/g and for Ge-68/Ga-68 is 10 Bq/g [5, 6]. For the Co-57 flood source, the MDA is 1.00 Bq/g so the sensitivity is adequate. On the contrary, for the Ge-68/Ga-68 line source the MDA is 812 Bq/g. So, the technique for the clearance of flood sealed radioactive sources of Co-57 is accurate and sensitive enough and for Ge-68/Ga-68 line sources is accurate but the sensitivity should be improved. It should be mentioned that it is possible to conduct future clearance measurements for Ge-68/Ga-68 line source without the source shielding structure for higher sensitivity. Furthermore, future positron simulation and use of the annihilation peak will improve the technique sensitivity.

Table 1. Minimum Detectable Activities and determined activities for different acquisition time, the formula used is $MDA = \frac{3 \cdot \sqrt{Background}}{(efficiency)\cdot(acquisition\ time)\cdot(fraction\ yield)}$.

Energy	Duration	Detection Limit (Bq/g)	Activity (MBq)
		Co-57	
122.06	7h	0.532 ± 0.001	1.569 ± 0.004
136.47	7h	3.89 ± 0.01	1.523 ± 0.004
692.03	7h	230.8 ± 0.9	1.47 ± 0.01
122.06	2h	1.016 ± 0.003	1.553 ± 0.004
136.47	2h	7.34 ± 0.02	1.579 ± 0.005
692.03	2h	433 ± 2	1.58 ± 0.02
122.06	15min	2.874 ± 0.008	1.508 ± 0.004
136.47	15min	20.72 ± 0.06	1.496 ± 0.005
692.03	15min	1224 ± 5	1.95 ± 0.05
		Ge-68/Ga-68	
1077.4	7h	309 ± 4	1.23 ± 0.01
1077.4	1h	812 ± 10	1.31 ± 0.02
1077.4	15min	1540 ± 19	1.32 ± 0.02

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