MEASUREMENT TECHNIQUE FOR CHARACTERIZATION AND RECORDING OF SPENT ^{57}Co AND $^{68}Ge/^{68}Ga$ SEALED RADIOACTIVE SOURCES

by

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The present work concerns the development of a technique for activity determination of $^{68}Ge/^{68}Ga$ and ^{57}Co disused radioactive sources. This technique aims to determine the activity of these sources by measuring and Monte Carlo simulation using the MCNPX code. Therefore, efficiency calibrations of the 3 3 NaI(Tl) detector for specific source geometries were carried out. Spectrums for two types of disused radioactive sources were collected for different measurement times. The characteristic gamma rays of a flood source containing ^{57}Co and a line source containing $^{68}Ge/^{68}Ga$, were used. In case of $^{68}Ge/^{68}Ga$, the annihilation peak of 511 keV was also used considering the disused radioactive sources as a positron emitter. Sources of the mentioned types with certified nominal activities were used to validate MCNPX models. For the ^{57}Co source, 15 minutes measuring time was adequate for activity determination, and 2 hours measuring time provided adequate sensitivity, at the level of general clearance. For $^{68}Ge/^{68}Ga$ line source, 15 minutes measurement was adequate for activity determination by using the 511 keV from the annihilation of 1899.1 keV positrons.

Key words: gamma-ray spectrometry, MCNPX, clearance, sealed radioactive source characterization

INTRODUCTION

Nowadays, various disused radioactive sources (DRS), like ^{57}Co and $^{68}Ge/^{68}Ga$ were originally used to precisely calibrate the nuclear medicine systems. After the end of their use at the hospitals, these sources must be handled and kept in storage until they meet the general clearance criteria. In many cases, their current activity cannot be estimated due to the lack of source certificates which include their nominal activity and the reference date. A technique was developed for the radiological characterization and recording of $^{68}Ge/^{68}Ga$ line and ^{57}Co flood DRS. This technique is based on non-destructive gamma spectrometry measurement and Monte Carlo simulation using the MCNPX code. The detector efficiency for the specific source-detector geometry was evaluated. In order to validate the technique, ^{57}Co and $^{68}Ge/^{68}Ga$ sources of these geometries and certified activities were used. The $^{68}Ge/^{68}Ga$ certified source was measured inside its shielding due to the high dose rate.

In previous papers [1, 2], a preliminary semi-empirical method was developed by using the characteristic gamma rays from the decay of ^{57}Co and $^{68}Ge/^{68}Ga$ and source-detector Monte Carlo simulations for clearance of DRS.

In this work, the line source of $^{68}Ge/^{68}Ga$ inside its shielding was simulated as a positron source and the annihilation peak of 511 keV was used to determine the source activity.

According to a published study [3], an important requirement in the spectrometry of positron emitters, based on the 511 keV full-energy peak, is complete annihilation within the materials that constitute the source. Positrons are generally stable in a vacuum but rapidly thermalize and annihilate with electrons in materials. Their mean lifetime is typically only a few nanoseconds [4]. Such spectrometry is often performed in cases where a particular radionuclide has significant $\beta+$ emissions but lacks other characteristic gamma-lines suitable for quantitative radioactivity assessment.

MATERIALS AND METHODS

Detection system and calibration sources

The gamma spectrometry system consists of the following main parts:

– Detector unit: A Bicron Monoline scintillation detector NaI(Tl) (Model 3M3/3) with a 3' 3' crystal

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in a thick Al housing, covered with a white reflector, including a 14-pin connector, an internal magnetic/light shield, and a photomultiplier tube.
- Electronics and the acquisition unit consist of a digital signal processing unit (Osprey Digital Tube Base MCA) and a high voltage supply system (850 V)
- GenieTM 2000 spectroscopy software for spectrum acquisition [5]

Firstly, the efficiency calibration of the detector was carried out inside the shielded chamber at the laboratory, by using standard calibration sources. This practice is well known and has been used by others [6, 7]

The experimental data for the efficiency calibration of the detector at the laboratory were obtained from a multiple gamma-ray standard volume source of 1.5 gcm^{-3} epoxy material density and a ^{40}K volume source with a similar density and the same geometry. The main features of the sources are shown in, tab. 1.

The experimental full-energy peak efficiency (E_{exp}) is the ratio of the counts N detected in a peak, to the number of photons emitted by the source [8]. The formula is $E_{exp} = N/(A\ t\ P\ )$, where N is the number detected counts, A – the source activity, t – the acquisition time, and P_{γ} – the probability of emission of the particular gamma-ray being measured (peak energy abundance). The main factors that may affect the efficiency calibration curve are the source-to-detector distance, the geometry of the source, the absorption within the source, the random summing at a high counting rate, and true coincidence summing at close geometry for specific radionuclides [8].

Experimental efficiency calibration was performed for the characteristic gamma-rays of, ^{137}Cs, ^{60}Co, and ^{40}K, tab. 1, with the volume sources adapted on an acetyl holder which was positioned in a vertical direction along the symmetry axis of the detector. The acquisition time was 1 hour and 40 minute. The dead time during the measurements was less than 2 %.

The MCNPX simulations were performed for efficiency calibration in an energy range of 45-1460 keV, including the characteristic gamma-ray energies of the volume sources, tab. 1. The detector was simulated as a NaI cylinder of 7.62 cm in diameter and 7.62 cm in height.

For the efficiency evaluation, the *F8* tally was used, which describes the distribution of energy for the pulses created in a detector by gamma radiation. The experimental efficiencies for the characteristic gamma-rays of ^{137}Cs and ^{40}K were used to evaluate the efficiency calibration curve, because of the possibility of true coincidence summing for the ^{60}Co peaks.

Table 1. Features of the gamma-ray certified volume sources

Source density [gcm^{-3}]	Isotopes	$T^{1/2}$ [d]	Reference activity [kBq]	Reference date
1.5 0.1	^{137}Cs	11001	2.66 2.9 %	1/3/2007
	^{60}Co	1923	3.32 2.9 %	
1.5 0.1	^{40}K	456 10^9	1.3 2 %	

Monte carlo simulations for the medical radioactive sources measurements

For the evaluation of the source-detector efficiencies, simulations were performed using the MCNPX code. The $^{68}Ge/^{68}Ga$ line source was simulated as a ceramic cylinder of height 16.3 cm and radius 0.07 cm inside its cylindrical Pb shielding, fig. 1. The source shielding was of height 37 cm, radius 2.85 cm and thickness 1.52 cm. The shielding of the source was placed at a distance of 25 cm from a 3 3 NaI detector geometrical center. Lead cylindrical shielding of height 18 cm, radius 7.5 cm, and thickness 3.3 cm, was placed around the detector to reduce the background radiation.

The ^{57}Co flood source was simulated as a rectangular parallelogram of length 61 cm and height 41.9 cm fig. 1. The flood source was placed at a distance of 47 cm from a 3 3 NaI detector geometrical center.

For the characteristic gamma-rays detection, the sources were simulated as gamma-ray emitters. When the annihilation peak 511 keV was used, the $^{68}Ge/^{68}Ga$ line source was simulated as a positron emitter.

Determination of the activity for the medical radioactive sources

Gamma-ray spectra were taken for different acquisition times. A 3' 3' NaI(Tl) detector was used to acquire the spectra.

Two types of DRS were measured:
- a line source containing $^{68}Ge/^{68}Ga$, fig. 1,
- a flood source containing ^{57}Co, fig. 2.

The uncertainty of the certified activities for these sources is 15 %.

For the $^{68}Ge/^{68}Ga$ line source, the acquisition times were 7 hours, 1 hour, and 15 minutes. For the ^{57}Co flood source, the acquisition times were 7 hours, 2 hours, and 15 minutes. The ^{68}Ge decays by electron capture to ^{68}Ga. The ^{68}Ga is mainly a positron emitter (89.1 %), and leads to ^{68}Zn. Then, due to the decay of the excited state of the daughter nucleus ^{68}Zn a gamma-ray of 1077 keV energy (2.93 %) is emitted [9]. The ^{57}Co decays by electron capture to the excited state of ^{57}Fe. Subsequently, gamma 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 ^{57}Fe [9]. The software package SPECTRW [10] was used to analyze the characteristic gamma-ray peaks in the resulting spectra. Also, the same software package was used in order to analyze the two overlapping photopeaks, 122 keV and 136 keV, emitted from the ^{57}Co source, fig. 3.

The determination of the activity for the DRS was carried out by using the following energy peaks:
- 1077 keV and 511 keV from the annihilation of 1899.1 keV positrons, for the $^{68}Ge/^{68}Ga$ line source
- 122 keV for the ^{57}Co flood source.

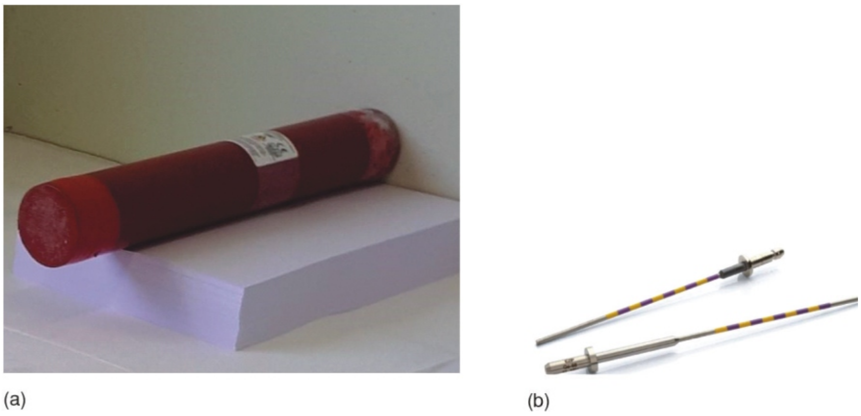

Figure 1. The ^{68}Ge/^{68}Ga line source with (a) and without (b) shielding

Figure 2. The ^{57}Co flood source

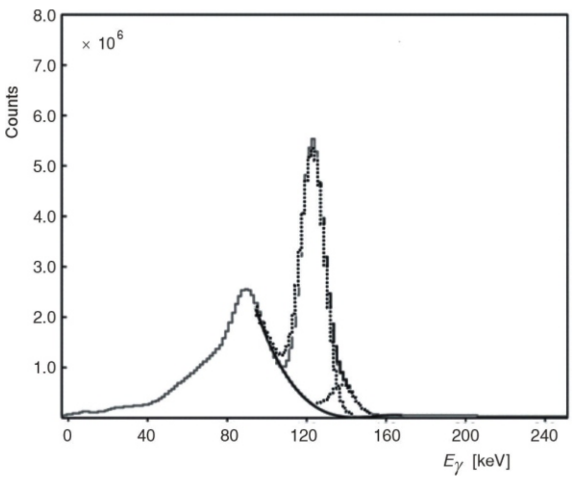

Figure 3. The ^{57}Co flood source spectrum for 2 hours measurement; the overlapping of 122 keV and 136 keV photopeaks is represented by solid line; dash line represents these two peaks separately

The detector main axis of symmetry and the flat surfaces of the sources were placed perpendicular to each other. In order to achieve an acceptable dead time (less than 5 %), the sources were placed at appropriate distances from the detector. It should be mentioned that the 511 keV energy peak was not analyzed as a Gaussian distribution. The Doppler effect during the annihilation of the positrons was taken into consideration, fig. 4. Therefore, the net counts were calculated by subtracting the background, configured as a trapezoid, fig. 4(b), from the total number of counts.

RESULTS AND DISCUSSION

The theoretical efficiency curve for the standard volume calibration source measurement inside the shielded chamber at the laboratory is presented in fig. 5. The energy range of the curve was chosen in order to include all the characteristic gamma rays of the DRS, as well as the 511 keV annihilation peak.

This work aims to characterize and record DRS without certificates, used for the calibration of nuclear medicine systems. A semi-empirical gamma spectrometry technique was developed by MCNPX simulations for ^{68}Ge/^{68}Ga line sources and ^{57}Co flood sources activity determination. For the evaluation, certified DRS of the same types with certificates were used. The determined activities are displayed compared to the nominal activities in figs. 6 and 7. The activities for both sources are in agreement with the nominal activities.

The results of the study show that an acquisition time of 15 minutes provides adequate statistics for the ^{57}Co flood sources activity determination, tab. 2. Nevertheless, a measurement of 2 hours is needed to achieve enough sensitivity. The minimum detectable activity (MDA) is 0.85 Bqg^{-1} for 2 hours of measurement, tab. 2, which is lower than the general clearance criterion of 1 Bqg^{-1} for ^{57}Co [11-13].

A 15 minutes measurement is adequate for the ^{68}Ge/^{68}Ga line sources activity determination regardless of whether we used the 511 keV or the 1077.4 keV energy peak. Nevertheless, by using the 511 keV annihilation peak, considering the DRS as a positron emitter, the MDA is about 10 times lower. According to the results shown in tab. 3, the MDA for measurement of

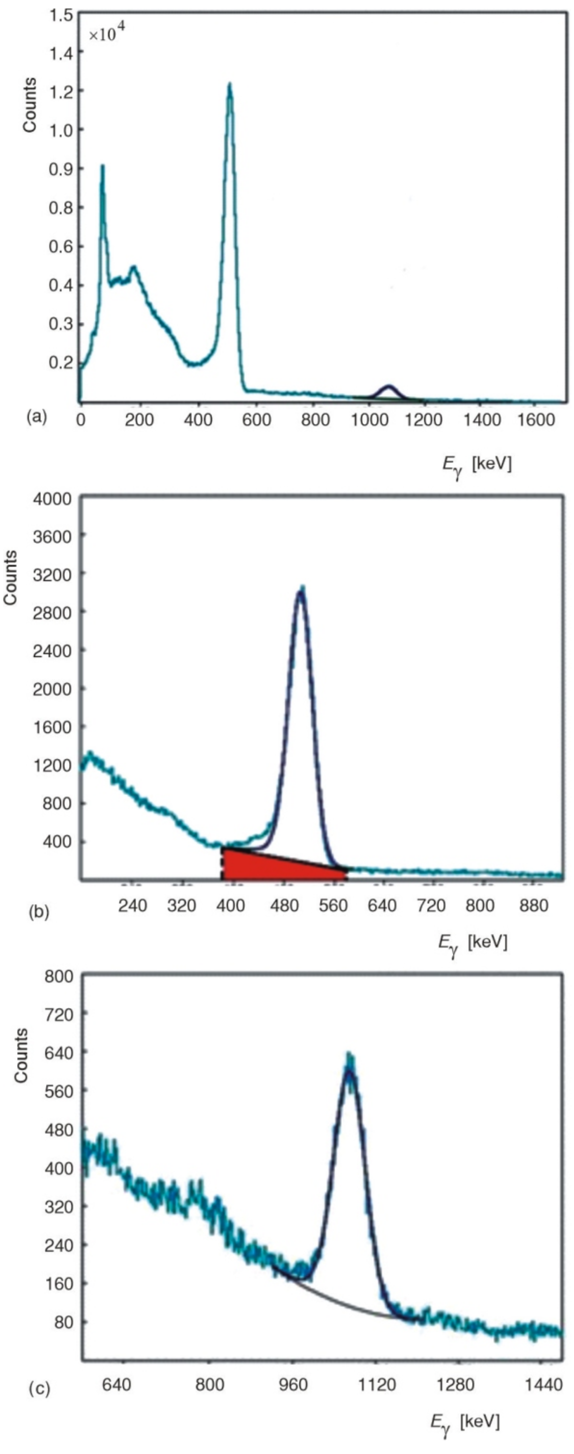

Figure 4. (a) The ^{68}Ge/^{68}Ga line source spectrum for 1 hour measurement, (b) annihilation peak with Doppler broadening and background shaped as a trapezoid, and (c) 1077 keV energy peak

1 hour for the annihilation energy peak of 511 keV is 90.37 Bqg^{-1}, which is higher than the clearance criterion of 10 Bqg^{-1} [11-13]. It should be mentioned that the ^{68}Ge/^{68}Ga source was located in lead shielding due to the high dose rate. Therefore, a much lower MDA will be achieved by measuring the line source without shielding in the future in case of clearance measurements.

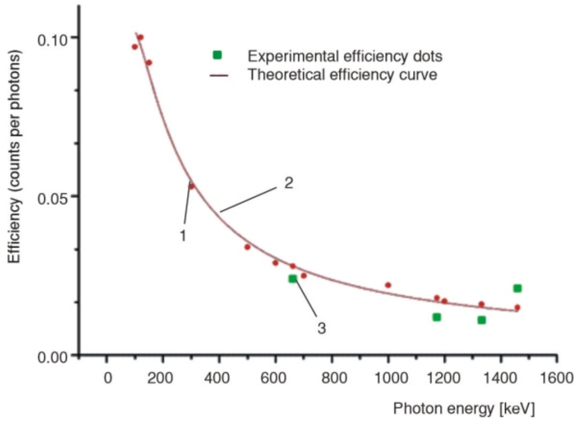

Figure 5. Theoretical efficiency values from MCNPX simulations (1), the corresponding calibration curve obtained by fitting a hyperbolic function to the theoretical values (2) as well as the experimental efficiency values (3)

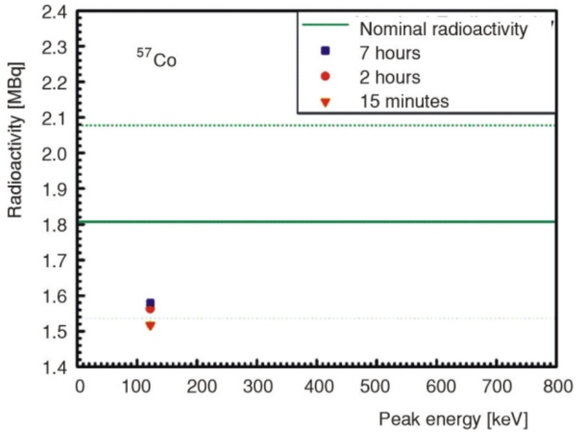

Figure 6. The ^{57}Co flood source determined activities for different acquisition times in comparison to the nominal activity; the dotted lines indicate the range of values within one standard deviation of the source-certified activity

CONCLUSIONS

Efficiency calibration of the detector was carried out inside the shielded chamber at the laboratory by using standard volume sources in order to ensure that the detection system works properly.

A semi-empirical gamma spectrometry technique was developed for the determination of the activities of 2 types of DRS (^{57}Co flood source and ^{68}Ge/^{68}Ga line source) which are used for the calibration of nuclear medicine systems. The technique is effective and accurate for the characterization and recording of these types of sources by 15 minutes measurement. Furthermore, clearance measurements can be carried out for spent DRS. The MDA for the ^{57}Co flood source by using the 122 keV energy peak is 0.85 Bqg^{-1} in 2 hours measurement. This is lower than the general clearance criterion of 1 Bqg^{-1} . Regarding the ^{68}Ge/^{68}Ga line source located in a lead shielding because of the high dose rate, the MDA for the characteristic energy peak 1077.4 keV is 309 Bqg^{-1} in 7 hours measurnement, which is much higher than the general

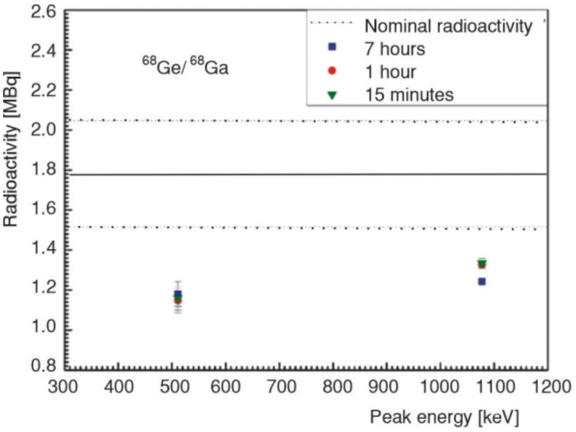

Figure 7. The ^{68}Ge/ ^{68}Ga line source determined activities for different acquisition times in comparison to the nominal activity; the dotted lines indicate the range of values within one standard deviation of the sources-certified activities

Table 2. The ^{57}Co flood DRS determined activities and MDA by using 122.06 keV energy peak for different acquisition time

Measurement time	Activity [MBq]	MDA [Bqg^{-1}]
7 hours	1.569 0.004	0.45
2 hours	1.553 0.004	0.85
15 minutes	1.508 0.004	2.42

Table 3. The ^{68}Ge/^{68}Ga lie DRS determined activities and MDA for different acquisition time

Energy [kev]	Measurement time	Activity [MBq]	MDA [Bqg^{-1}]
511	7 hours	1.18 0.06	33.4
1077.4	7 hours	1.23 0.02	309
511	1 hour	1.15 0.06	90.4
1077.4	1 hour	1.31 0.02	812
511	15 minutes	1.16 0.06	174
1077.4	15 minutes	1.32 0.02	1540

clearance criterion. Nevertheless, MDA for measurement of 1 hour by using the energy peak of 511 keV from the annihilation of 1899.1 keV positrons is 90.4 Bqg^{-1}. Although this MDA is higher than the general clearance criterion of 10 Bqg^{-1}, a much lower MDA can be achieved by measuring the line source without shielding in the future for direct release.

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AUTHORS' CONTRIBUTIONS

The first author is in responsible of the manuscript's content, he performed the efficiency calibration of the detector, checked all the codes and calculations, and wrote the manuscript. The second author developed the MCNPX simulations for the line and flood sources and carried out the preliminary calculations. The third author analyzed the gamma-ray spectra of the line and flood sources with the SPECRW software. The fourth author installed the measurements set-up and supported the development of the MCNPX models. The fifth author co-supervised the project work, and the sixth author supervised and controlled the project work.

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ТЕХНИКА МЕРЕЊА ЗА КАРАКТЕРИЗАЦИЈУ И РЕГИСТРОВАЊЕ ИСТРОШЕНИХ ^{57}Co И ^{68}Ge/^{68}Ga ЗАТВОРЕНИХ РАДИОАКТИВНИХ ИЗВОРА

Рад се бави развојем технике за одређивање активности одложених ^{68}Ge/^{68}Ga и ^{57}Co радиоактивних извора. Техника има за циљ да одреди активност ових извора мерењем и Монте Карло симулацијом коришћењем MCNPX програма. Због тога су извршене калибрације ефикасности 3 3 NaI(Tl) детектора за специфичне геометрије извора. Спектри за две врсте утрошених радиоактивних извора прикупљени су за различита времена мерења. Коришћено је карактеристично гама зрачење из извора који садржи ^{57}Co и линијског извора који садржи ^{68}Ge/^{68}Ga. У случају ^{68}Ge/^{68}Ga, такође је коришћен анихилациони пик од 511 keV, с обзиром на ислужене радиоактивне изворе као емитере позитрона. За валидацију MCNPX модела коришћени су извори поменутих типова са сертификованим номиналним активностима. За извор ^{57}Co, време мерења од 15 минута било је довољно за одређивање активности, а два сата мерења је обезбедило адекватну осетљивост, на нивоу општег клиренса. За линијски извор ^{68}Ge/^{68}Ga, мерење од 15 минута било је адекватно за одређивање активности коришћењем 511 keV од анихилације позитрона од 1899.1 keV.

Кључне речи: спектрометрија гама зрачења, MCNPX програм, клиренс, карактеризација затвореног радиоактивног извора