OPTIMIZATION APPROACH OF GAMMA SPECTROMETER MEASUREMENTS FOR ACCURATE RADIOACTIVE MATERIALS CHARACTERIZATION

by

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Scientific paper
https://doi.org/10.2298/NTRP2303187G

The materials characterization based on gamma measurements is one of the main features for verifying the radioactive materials for safeguards purposes. This work focuses on the optimization approach in order to find solutions that include some influence parameters, which should be taken into consideration during measurements. The considered samples in our work are certified reference nuclear materials of chemical composition (U_3O_8) with different enrichment ratios ranging from 0.31 wt.% to 4.46 wt.%. The gamma spectrometer based on a planar high-purity germanium detector of high-resolution was used in the current study. The study of various setups was experimentally carried out for different cans at different positions for different energy lines. The verified parameters influencing the characterization of the measured samples have been estimated. This approach for the measurement setup of the measured gamma spectra has been successfully explored to be very affected by different parameters such as the source to detector position and the various enrichment ratios. Consequently, the verified and characterized samples could be estimated based on this optimization approach.

Key words: gamma spectrum, uranium enrichment, high-purity germanium detector, count rate

INTRODUCTION

Estimating uranium enrichment is a crucial aspect of radioactive measurements. Furthermore, it is thought to be a crucial component of safeguards duties for checking nuclear materials and ensuring that they are controlled for peaceful nuclear operations.

The necessary methods for safeguards purposes should not affect the concentrations of nuclear materials; for example, non-destructive analysis (NDA) can be carried out using detection techniques that rely on gamma-ray spectrometry to characterize and measure the nuclear materials could be used [1, 2]. The uranium enrichment meter mode is the oldest and most widely used of these several techniques. The proportionality link between the number of counts per specific time at the 185.7 keV energy line from ^{235}U decay and enrichment between an unknown estimated sample and a calibration standard is used to suggest this mode. Additionally, it requires a single calibration test using a standard of known enrichment and thickness of the container wall [3]. The predicted sample differences in thickness of the container wall and geometric shape make it difficult to meet all the necessary calibration requirements in the measurement field. Additional self-calibration-based techniques have been proposed to get rid of/resolve these issues. A relationship is established between the intensity ratios of the self-fluorescence X-ray situated within the UXK spectral region and the ^{235}U enrichment ratios of the known standard through self-calibrating analysis based on the calibration curve. To assess ^{235}U enrichment, one can use intensity ratios such as 94.6 keV and 98.4 keV peaks produced by the ^{231}Th decay product of ^{235}U in the UXK spectral area to 92.6 keV gamma-rays from the ^{234}Th decay product of ^{238}U. Due to the energy of gamma lines being so close together, this analysis does not require calibration [3-5]. The X-ray and gamma-ray spectrometry techniques have been developed for measuring the uranium enrichment ratio based on the previously mentioned analysis. In particular, the simplicity of self-calibrating measurements of radioactively assessed samples containing uranium was demonstrated by the multi-group gamma-ray analysis for uranium (MGAU) [6-9]. Additionally, when a suitable detection system of high resolution is used, this computerized tool has the potential to produce fast assay and accurate results, especially in nuclear safeguards inspections at nuclear facilities. Results for measured depleted and highly enriched

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samples are thought to be provided by the MGAU approach within a relative variance of up to 2 %, as studied [3, 8]. Its measuring limitations include requiring the assessed sample to be no thicker than 6 mm and ensuring that the yields of the uranium isotopes balance with each other, even if it offers a fast and satisfactory performance. Additionally, in order to use its enrichment meter approach, a high-resolution detection system that has been previously calibrated using standard nuclear materials with known enrichments and similar geometries to the estimated items is required [9]. Even for long measuring times, this technique may not detect or produce high errors for natural and depleted uranium as well as samples with low uranium quantities. If so, it is possible that this method is not entirely appropriate or trustworthy to use for enrichment computations [10, 11]. Therefore, it is important to look for different strategies or methods to get beyond these restrictions in order to improve performance and outcomes. The Monte Carlo simulation using the MCNP code is one of these substitute methods [12]. In general, it is a useful tool for modeling intricate geometries and NDA devices [13-16]. Furthermore, it can provide the necessary cross-sectional data for computations involving the transit of electrons, neutrons, and photons [17]. As a result, the MCNP code is an appropriate tool for achieving calibration and accurately assessing the detecting system's full energy peak efficiency (FEPE) [18-20]. The MCNP code is appropriate for nuclear safeguards activities involving uranium metal samples based on the findings of FEPE, and its findings are consistent with the measurement [21]. It can be used to investigate how various parameters affect the measurements; such as the size of the detector, the source-detector (S-D) position, the thickness of the shielded container, the net area under the gamma peak, the amount of uranium in the sample, *etc*. [22-26].

The current study investigates the factors and parameters influencing the characteristics of the measured gamma spectra based on experimental measurements of some standard reference materials of different enrichments and sizes. It was concluded that the considered influencing parameters can affect the characteristics of the measured gamma spectra.

Egypt and the IAEA have a safeguardss agreement that applies to all certified reference samples that are used.

EXPERIMENTAL SETUP AND TECHNIQUES

Utilized materials

A set of certified reference materials (CRM) of uranium oxide (U_3O_8) consist of five samples of depleted, natural and low enriched uranium of different abundance ratios, ranging from 0.31 wt.% to 4.46 wt.% have been utilized for the measurements. Each sample has a distinct enrichment ratio while having a cylindrical shape. In the current study, the samples were organized according to their ID numbers, CRM1, CRM2, CRM3, CRM4, and CRM5. The samples were packed in an aluminum cylindrical container with an internal diameter of 70 mm and an exterior diameter of 80 mm, as well as a height of 89.9 mm. Additionally; all samples have an nuclear materials filling height of (20.8 0.5) mm, with the exception of the CRM5 of abundance 4.46 wt.%, which has a height of (15.8 0.5) mm. There are 200.10 g of U_3O_8 powder in each sample. The samples that were used are disclosed in compliance with Egypt and the IAEA's safeguardss Agreement.

Experimental configuration and system set-up

In the current work, an high-purity germanium (HPGe) detector, based on a planar crystal of model GL0515R (Canberra manufacture) was used. Energy resolution of 0.540 keV was measured at 122 keV (^{57}Co). Additionally, a liquid nitrogen (5L) cooling system and cryostat (model 7905 SL-5) are integrated with the detector. In order to store events according to a single parameter (pulse height), the gamma spectrometer utilized in this experiment is coupled with traditional electronic components to create a portable Inspector multi-channel pulse-height analyzer (MCA). The energy window was set to 8 k channels and the detector was set to a high voltage (–2500 V). Genie 2000 (GAA) and MGAU software are included with the Canberra system to help with spectrum acquisition and data analysis for the purpose of determining ^{235}U enrichment. As shown in fig. 1, the samples were positioned axially with respect to the detector crystal and were measured under various conditions.

In order to get a sufficient count rate with acceptable counting statistics, the experiment was conducted three times, with each run lasting between 1800 seconds and 7200 seconds between measurements. In order to obtain high-quality spectra of valuable shapes, the measuring position of all objects that were analyzed was chosen to ensure accurate count rates at the conspicuous high-intensity gamma energy of 185.7 keV. In addition, the dead time was maintained below 1 % in order to reduce the impact of genuine coincidences. Additionally, by adjusting the sample to the detector window distance as much as feasible, the measurements were performed as close to the ideal conditions as possible. Additionally, as shown in fig. 2, a broad range of energy calibration has been used to obtain acceptable uranium spectra.

RESULTS AND DISCUSSION

The results of the uranium enrichment measurements of the standard reference materials

As illustrated in tab. 1, for the estimated certified reference materials, we can notice that the results from uranium samples based on the MGAU method for the

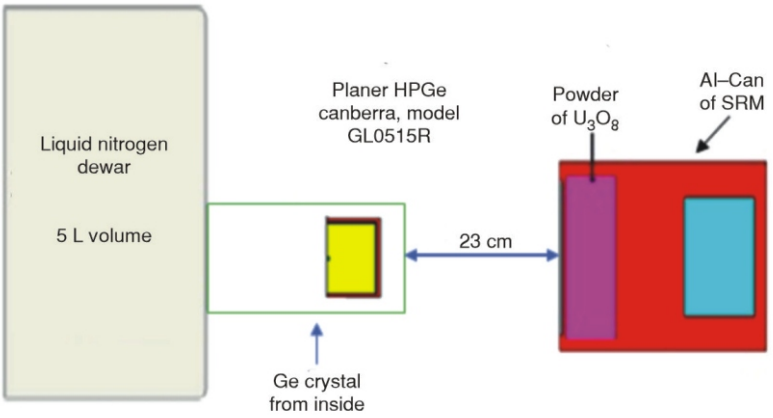

Figure 1. Geometrical set-up of the measured certified nuclear materials

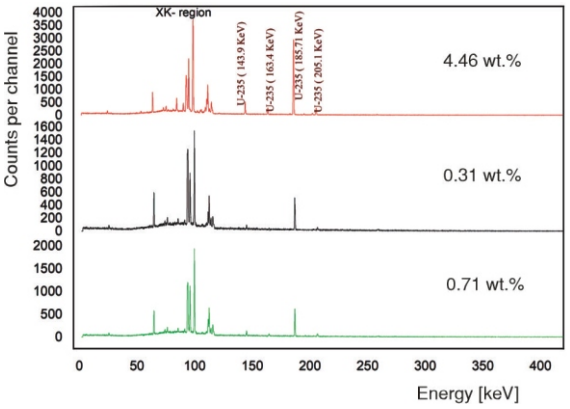

Figure 2. Gamma spectra of U_3O_8 oxide samples with various enrichment 0.31 wt.%, 0.71 wt.%, and 4.46 wt.% in the energy region of 0-400 keV

samples CRM1, CRM2, and CRM3 are relatively far from the certified values. Thus, the MGAU results are not in agreement with the certified reference values. Moreover, MGAU enrichment analysis is accompanied with a relative high uncertainty of 6.85 %, 5.76 %, and 4.22 %, for the depleted (0.31 wt.%), natural (0.71 wt.%) and low enriched samples (1.94 wt.%), respectively. These uncertainties can be explained so that the uranium bearing samples containing a low amount of uranium result in non-sufficient detected photons and so low-quality gamma spectra of poor statistics. Moreover, the MGAU's algorithms analysis is a function of the detector resolution, in which the MGAU required a certain detector with a relatively high resolution. In addition to the abovementioned, the self-attenuation of the estimated samples and the attenuation due to the container wall of the sample can also influence the results of the MGAU analysis.

On the other hand, for the certified samples CRM4 and CRM5 of relatively high uranium enrichment, the MGAU analysis gives reasonably acceptable results within the associated relative uncertainty. Furthermore, it is very obvious that the natural sample CRM2 of enrichment 0.71 wt.% has the maximum relative bias of 9.85 % in comparison to the low enriched sample CRM3 of enrichment 1.94 wt.%. We can justify/explain the high relative bias of the results of Uranium enrichment that the MGAU's analysis is based on the ^{235}U amount/isotopic ratio, in which the build-up of the decay products begins to interfere with the analysis. Also, it is shown that the MGAU analysis is a function also of the sample concentration, in which for samples containing a large uranium amount, the associated result uncertainties tend to reduce and be close to the certified values. So, the associated uncertainties of the MGAU analysis for enriched samples CRM4 and CRM5 have been decreased to be 3.96 % and 2.24 % respectively.

The parameters influencing the measured gamma spectra

Sample to detector position affecting the full energy peak efficiency

An obtained correlation between the counted gamma photons and the S-D position as one of the parameters affecting the measured gamma spectra is illustrated in figs. 3-5. It is clear that the FEPE is attenuated and has high values at the near distances and low

Table 1. The measured ^{235}U enrichment, and the recorded count rate of the HPGe-detector

Sample	Filling height [mm]	Mass of ^{235}U [g]	E_n [wt. %] enrichment	Relative uncertainty [%] [1]	Relative bias [%] [2]
CRM1	20.800 ± 0.5	0.526	0.321 0.022	6.853	–3.426
CRM2	20.800 0.5	1.205	0.780 0.045	5.769	–9.859
CRM3	20.800 0.5	3.292	2.084 0.088	4.222	–7.422
CRM4	20.800 0.5	5.006	2.925 0.116	3.965	0.847
CRM5	15.800 0.5	7.567	4.587 0.103	2.245	–2.847

[1]Relative uncertainty = (the error of the measured value / the measured value) 100
[2]Relative bias = [(declared value- measured value)/declared] 100

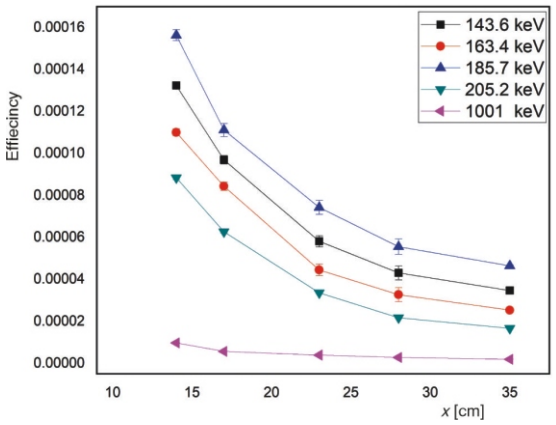

Figure 3. Full energy peak efficiency of the measured CRM1 of 0.31% at a different S-D position and different energy lines

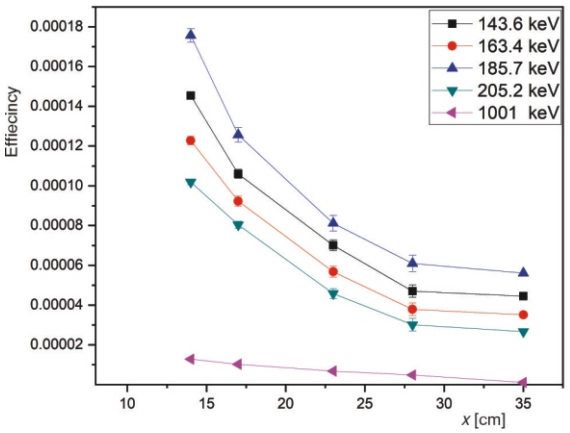

Figure 4. Full energy peak efficiency of the measured CRM2 of 0.71 % at a different S-D position

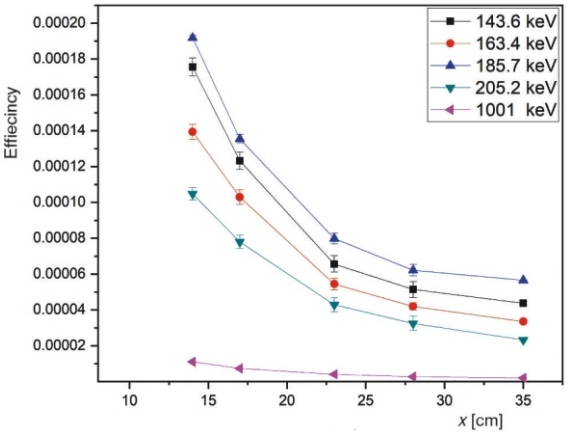

Figure 5. Full energy peak efficiency of the measured CRM5 of 4.46 % at a different S-D position

values for the far distances, so the FEPE has an inverse relation with the S-D distance as it is an important parameter that affects the results from uranium samples of the MGAU analysis as depicted in fig. 6.

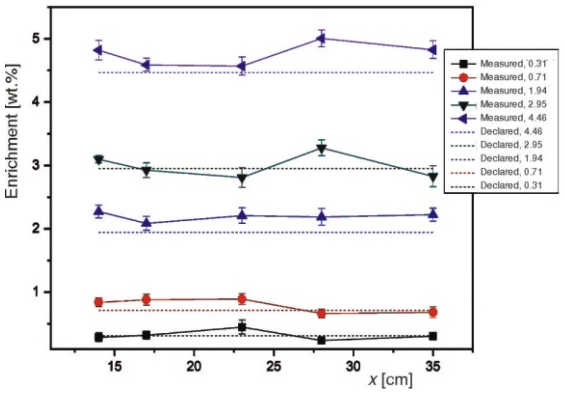

Figure 6. Relation of the enrichment at a different S-D position for different samples

While the following equation defines FEPE as the ratio of the number of recorded or counted photons that reach the detector to the number of photons emitted by the source

$$\varepsilon \quad \frac{C_p}{E_p} \tag{1}$$

A gamma-ray beam of emitted photons E_p will attenuate and record at the detector as counted photons C_p, which can be determined using Beer-Lambert's law, when it passes through an S-D position, x, and linear attenuation coefficient, μ. Thus, the correlation between the S-D position and the linear attenuation coefficient of photons in gamma rays is

$$C_p \quad E_p e^{\ \mu x} \tag{2}$$

Using eq. (1) in eq. (2), a coefficient can be acquired as indicated in eq. (3), in which the efficiency has an inverse relation with the attenuation coefficient

$$\varepsilon \quad e^{\ \mu x} \tag{3}$$

Sample to detector position at different energies influencing the gamma counts

One of the crucial factors affecting the recorded gamma spectra is also shown in figs. 7-9, where it is shown that, for the confirmed certified reference material samples, the total number of counts is an inverse function of the S-D position. Moreover, it is also the enrichment's inverse function. Thus, there may be a relationship between the S-D position and uranium enrichment. An increase in the number of photons released means that there is a greater chance of counting the photons that interact in the detector's active volume, which produces sufficient count rates quickly enough for the analysis of gamma spectra. As illustrated in figs. 7-9, ordering of count rates for an energy peak of 1001 keV from ^{238}U is changed as the activity concentration of ^{235}U increases for more and more enriched samples while the ^{238}U activity concentration remains unchanged. Therefore, under ideal circum-

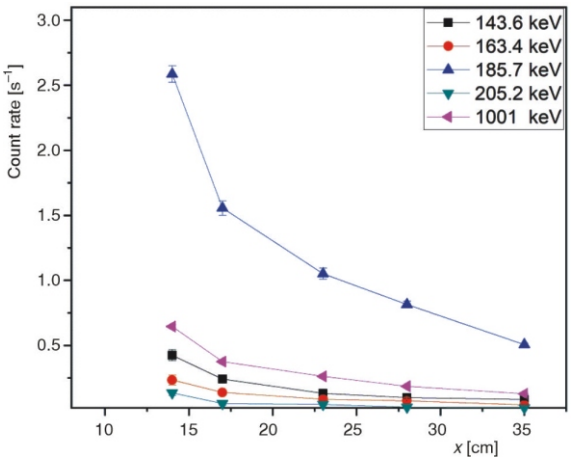

Figure 7. Relation of the count rate of the measured CRM1 of 0.31 % at a different S-D distance and different energy lines

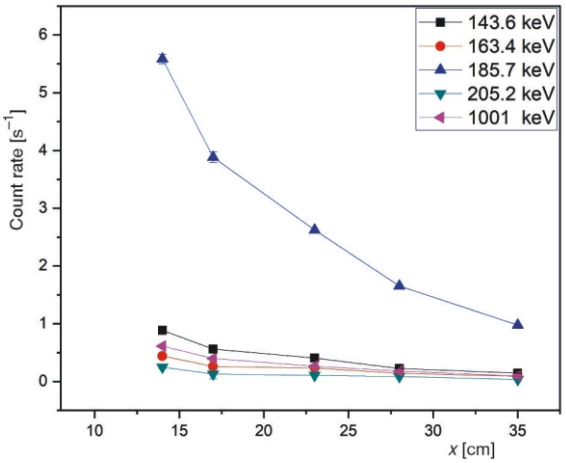

Figure 8. Relation of the count rate of the measured CRM2 of 0.71 % at a different S-D distance and different energy lines

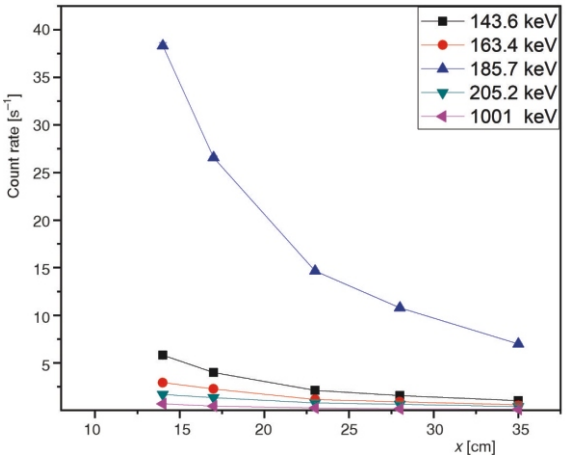

Figure 9. Relation of the count rate of the measured CRM5 of 4.46 % at a different S-D distance and different energy lines

stances, samples with a high concentration of uranium or samples with a significant amount of uranium could yield reliable results.

CONCLUSION

In this work, a proposed optimization approach concerned with the investigation of some parameters influencing the measurements of enrichment of the radioactive samples has been taken into consideration during the verification for safeguards purposes. Different samples of various enrichment ratios are assayed based on the HPGe detector system. The measurement results based on different positions for different can samples of different enrichment have shown that the investigated parameters can influence the verified samples. The study was experimentally carried out for different setup positions varying from 14 cm to 35 cm at different gamma energy lines. The performance of this approach has been successfully introduced, in which parameters influencing the measured gamma spectra, *i.e*., S-D position and enrichment ratios have been estimated. Thus, the verified and characterized samples may be accurately estimated based on this suggested approach.

AUTHORS' CONTRIBUTIONS

K. A. E. Gawad is responsible for the manuscript's content, wrote the manuscript and proposed the idea for the experiment. The measurements were carried out by M. H. Hazzaa, who performed the efficiency calibration of the detector, and carried out the preliminary calculations. S. E. Shaban analyzed the gamma-ray spectra and installed the measurement setup. All the authors analyzed the results and participated in the preparation of the final version of the manuscript.

ACKNOWLEDGMENTS

The authors wish to thank the Department of Nuclear safeguardss and Physical Protection, EAEA, Cairo, Egypt, for their support. Finally, we thank the EAEA, Egypt for funding this work.

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Received on September 23, 2023
Accepted on November 29, 2023

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ОПТИМИЗАЦИОНИ ПРИСТУП ГАМА СПЕКТРОМЕТРИЈСКИМ МЕРЕЊИМА ЗА ПРЕЦИЗНУ КАРАКТЕРИЗАЦИЈУ РАДИОАКТИВНИХ МАТЕРИЈАЛА

Карактеризација материјала заснована на гама мерењима један је од главних поступака за верификацију радиоактивних материјала у сврхе заштите. Овај рад фокусира се на приступ оптимизацији предложен у циљу проналажења решења која укључују утицајне параметре које треба узети у обзир током мерења. Разматрани узорци су сертификовани референтни нуклеарни материјали хемијског састава (U_3O_8) са различитим односима обогаћивања у распону од 0,31 wt.% до 4,46 wt.%. У раду је коришћен гама спектрометар заснован на планарном детектору германијума високе чистоће и високе резолуције. Експериментално је спроведено проучавање различитих лименки на разним позицијама, за различите енергетске линије. Процењени су верификовани параметри који утичу на карактеризацију мерених узорака. Приступ са подешавањем мерења измерених гама спектара успешно је истражен и установљен је утицај различити параметара, као што су положај извора и детектора и различити односи обогаћивања. Сходно томе, верификовани и окарактерисани узорци могу се проценити на основу овог приступа оптимизацији.

Кључне речи: гама спектар, обогаћење уранијума, HPGe детектор, брзина бројања