BENI-SUEF UNIVERSITY JOURNAL OF BASIC AND APPLIED SCIENCES 3 (2014) 63–67

Available online at www.sciencedirect.com

ScienceDirect

journal homepage: www.elsevier.com/locate/bjbas

# Validation for application of the Monte Carlo simulation code for $^{235}U$ mass content verification for large size samples of nuclear materials

M.S. El Tahawy [a], M.F. Eissa [b,*], H. Hamdy [b], M.A. Samei [a], Elham Elhakim [a], K. Abd El Gawad [a]

[a] Safeguards & Physical Protection Dept., E NRRA, Egyptian Nuclear and Radiological Regulatory Authority, P.O. Box 7551, Nasr City, 11762 Cairo, Egypt

[b] Physics Dept., Faculty of Science, Beni Suef University, Beni Suef, Egypt

ARTICLE INFO

*Article history:*
Received 22 December 2013
Received in revised form
16 February 2014
Accepted 17 February 2014
Available online 5 March 2014

*Keywords:*
Non-destructive assay
HPGe detector
Monte Carlo calculations
Nuclear material samples

ABSTRACT

In this work, a new semi- absolute non-destructive assay technique has been developed to verify the mass content of $^{235}U$ in the large sizes nuclear material samples of different enrichment through combination of experimental measurements and Mont Carlo calculations (version MCNP5). A good agreement was found between the calculated and declared values of the mass content of $^{235}U$ of uranium oxide ($UO_2$) samples. The results obtained from Mont Carlo calculations showed that the semi-absolute technique can be used with sufficient reliability to verify the uranium mass content in the large sizes nuclear material samples of different enrichment.



## 1. Introduction

A state with nuclear activities should establish a system capable of controlling all nuclear materials (NMs) under its authority that prevents any deviation or misuse of NMs from the planned peaceful activities. Continuous improvement of a measuring system is an essential mandate such controlling system. Measurements of NM using absolute methods could eliminate the dependency on NM standards, which are necessary for other relative or semi-absolute methods.

The determination of uranium fissile in samples is a key measurement for product and process control in fuel fabrication plants, waste characterization, tracking of nuclear materials issued in illicit trafficking and it is also very important in internationally nuclear safeguards inspections (Yücel and Dikmen, 2009; Anilkumar et al., 2007) In order to fulfill its national and international safeguards commitments,

* Corresponding author.
E-mail address: mfesms@yahoo.com (M.F. Eissa).

Peer review under the responsibility of Beni-Suef University

Production and hosting by Elsevier

http://dx.doi.org/10.1016/j.bjbas.2014.02.008

a state should establish and maintain a State System of Accounting for and Control of nuclear materials (SSAC) (El-Gammal, 2007).

One of the main functions of SSAC system is to verify the mass content of $^{235}$U in the state (Hamed et al., 2010).

To carry out inspection activities in state, the authorities are looking for rapid and easy methods of NM assay. These inspections must be possible, nondestructive assay, which favors γ-ray spectrometry over other methods like mass spectrometry. Moreover, it is desirable to get the accurate information in a short time of period without destroying the measured material (Anilkumar et al., 2007; El-Gammal, 2007).

The most widely employed methods of measuring $^{235}$U mass contents is carried out via measuring γ-ray line 185.7 keV (57.7%) of $^{235}$U full energy peak using gamma ray detectors (El-Gammal, 2007; Reilly et al., 1974; Matussek, 1985). Efficiency calibration of the detectors must be performed for each verified nuclear material. The characteristics of the verified NM and the experimental set up configuration must be taken into consideration while selecting the proper standard NM used for calibration. To obtain accurate results, standard NM with very similar characteristics to the verified samples has to be used. However, because suitable standard NMs are not always available, sometimes an appropriate calibration curve could not be constructed (Reilly et al., 1974).

The Monte Carlo method has been used to estimate the fissile content of a spent fuel assembly (Conlin and Tobin, 2011). The Intra-nuclear Cascade Monte Carlo (ICMC) code for transport of neutrons, protons, pions and heavy ions had been developed at Nuclear Physics Division .the code had been developed for low energy neutron transport using point wise cross section data below 20 MeV of neutron energy. Constructive Solid Geometry model, based on solid bodies, is adopted to construct geometry. A module for repetitive structure for lattice, core calculations in reactors and detector simulations is developed (Kumawat and Venkata, 2013). The Monte Carlo (version MCNP5) method can provide the solution regarding that issue (El-Gammal, 2007). Mont Carlo simulation technique, which is becoming progressively popular has been used by many authors since many years ago to simulate the process of gamma rays detection (Karamanis, 2003; Ewa et al., 2001; Lepy et al., 2001). It was used to calculate response characteristics of different detectors types at mono energetic and different gamma ray energies (Wainio and Knoll, 1966; Avignone, 1980; de Castro and Levesque, 1967; Hurtado et al., 2004; Meixner, 1974; Michel et al., 1986; Salgado et al., 2006; Varley et al., 1981; Fehrenbacher et al., 1996). It was also used for efficiency calibration of detectors, or directly through combination with experimental measurements (Karamanis, 2003; Ewa et al., 2001; Debertin and Grosswendt, 1982; Ashrfi et al., 1999; Rodenas et al., 2000; Kamboj and Kahn, 2003). The aim of this work is to verify the mass content of $^{235}$U-isotope in large sizes $UO_2$ samples through combination of experimental measurements and Mont Carlo calculations.

## 2. $^{235}$U Estimation

In order to cover the wide variety of different types and sizes of NMs existing in nuclear facilities, Research &Development work is directed toward developing techniques and methods that allow the verification of all existing NMs categories exist. The proposed technique is one of the R&D activities that could be applied to verify the NMs with different enrichment and sizes. Passive Non-destructive Assay Technique (NDA) was used to verify $^{235}$U mass content in large sizes NM samples using HPGe spectrometer in combination with the Monte Carlo calculations (version MCNP5). The MCNP calculations have been utilized to obtain the absolute full energy peak efficiency of an HPGe detector to estimate the nuclear material mass content. In the absence of high count rate, the relation between the net counting rate $C_R$ measured by HPGe detector under gamma ray line 185.7 keV (57.7%) of $^{235}$U isotope and its mass content (M) could be gives as (El-Gammal, 2007):

$$C_R = M \cdot S_a \cdot A_t \cdot \Omega_f \cdot \varepsilon_i \quad (1)$$

where $S_a$ is the specific activity of the measured gamma photons for this isotope (Bq $g^{-1}$), $A_t$ is the total attenuation correction factor due to attenuation in the radioactive material itself, matrix, container or clad, and all other media between the measured sample and the detector, $\Omega_f$ is the fractional solid angle of the sample subtended by the detector and $\varepsilon_i$ is the intrinsic full energy peak efficiency of the detector.

Equation (1) could be rewritten as:

$$C_R = M \cdot S_a \cdot \varepsilon_a \quad (2)$$

Where $\varepsilon_a$ is the absolute full energy peak efficiency which can be determined as:

$$\varepsilon_a = \varepsilon_i \cdot A_t \cdot \Omega_f \quad (3)$$

Eq. (2) shows that, the $^{235}$U-isotope mass content could be determined by measuring $C_R$ experimentally and calculating $\varepsilon_a$ using MCNP code.

The enrichment can be expressed as a weight fraction from the relation (Reilly et al., 1991):

$$E_w(\text{wt\%}) = \text{No. of grams}^{235}\text{U/No. of grams U} \times 100. \quad (4)$$

Where $E_w$ is the enrichment of $^{235}$U isotope, No. of grams $^{235}$U is the mass content of $^{235}$U-isotope and No. of grams U is the mass content of uranium element

## 3. Measurements

### 3.1. *Material and equipment*

In this study, two uranium materials samples, the first is162-3 (slightly enriched) and the second is UN-29121 (natural) consisting of nominal abundances 1.38% and 0.7204% of $^{235}$U isotope respectively were used for measurements. The samples contain $UO_2$ powder encased in carbon steel and tin cylindrical containers respectively, the characteristics and specifications of these samples are shown in Table 1.

A portable high-resolution gamma ray spectrometer is employed for the measurement which is composed of a high resolution planar Ge-detector [Canberra; model GL0515R with an active area of 540 mm$^2$, 1.5 cm height and 540 eV FWHM at 122 keV],a cryostat [model 7905 SL-5] with 5 L liquid nitrogen dewar, was used to cool the detector, a portable Inspector

**Table 1 – Material characteristics and specifications of the nuclear material samples.**

| Sample ID | NM ($UO_2$) | | | | Container | | | |
|---|---|---|---|---|---|---|---|---|
| | Weight (kg) | Height (cm) | Density ($g/cm^3$) | Enrichment (%) | Outer radius (cm) | Thickness (cm) | | Density ($g/cm^3$) |
| | | | | | | Lateral | Base | |
| 162-3 | 23.007 | 41.5 | 2.361 | 1.38 | 12.6 | 0.6 | 0.6 | 7.87 |
| UN-29121 | 25.014 | 15.4 | 2.501 | 0.72 | 14.6 | 0.2 | 0.2 | 7.31 |

Multi-channel Pulse-Height Analyzer [IMCA, Model 1200], for sorting and collecting the gamma-ray pulses coming from the main amplifier, an adjustable High Voltage Power Supply [HV-PS], provides a negative voltage of 2000 V which necessary for the operation of the detector, a laptop computer with spectroscopic software (Genie-2000) installed for data acquisition, display and analysis. Also, the MGAU [Ver.S507c] code for uranium enrichment estimation was installed on the system.

### 3.2. *Experimental setup*

The nuclear material samples were measured at different sample – detector distances (111.7 cm and 96.7 cm) with respect to the axis of symmetry of the detector as shown in Fig. 1.

## 4. Mont Carlo calculations

Mont Carlo code is an advanced simulation program containing all necessary cross-section data of neutron, photon and electron transport calculations (Keyser, 2004). During calculations photon is followed between collisions; its energy deposition is recorded, throughout its life to its death, until its energy is low enough to be neglected (1 keV).

Processes as coherent and incoherent scattering, photoelectric absorption and possibility of fluorescent are considered. The energy deposited in the active volume of the detector crystal can therefore be determined.

The MCNP5 version of the code has been used for modeling the detector response, since it contains a tally, F8 (absolute full energy peak efficiency of HPGe detector), which is specific for detector pulse height determination. The fraction of gamma-rays with certain energy absorbed in the detector active volume represents its absolute full energy peak efficiency at that energy. Accurate results in the calculated efficiency of the simulated detector could be obtained if accurate model for the experiment is developed then this the calculated efficiency is substituted in equation (2) to calculate the mass content of $^{235}U$-isotope.

The data provided by the detector's manufacturer were used to construct the MCNP input file. NM samples with specified characteristics were used to check the validity of the model. Then F8 card was used to calculate the absolute full energy peak of the detector at 185.7 keV gamma ray energy of the $^{235}U$-isotope which is the output of the MCNP input file. The simulation model was checked for reliability through performing measurements and calculations at different sample-detector distances.

Two MCNP input files were designed to calculate the detector absolute full energy peak efficiency at gamma ray energy. The calculations were performed for two nuclear material samples (enriched and natural) at two different distances.

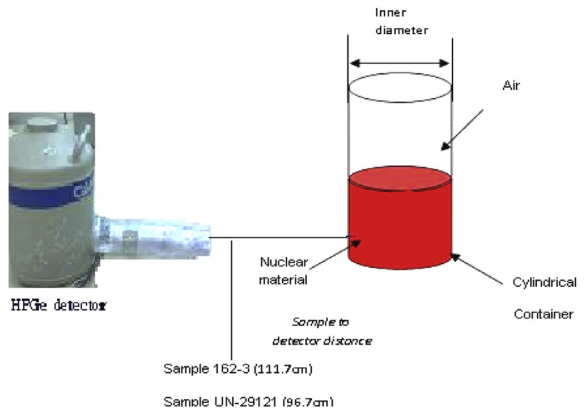


**Fig. 1 – The scheme of samples detector geometry.**

## 5. Results and discussion

The calculated absolute efficiency by using MCNP code and the net count rate measured by the detector were substituted into equation (2) to calculate the mass content of $^{235}U$ for the two samples at different distances. The calculated enrichment by using MGAU code and the mass content of uranium

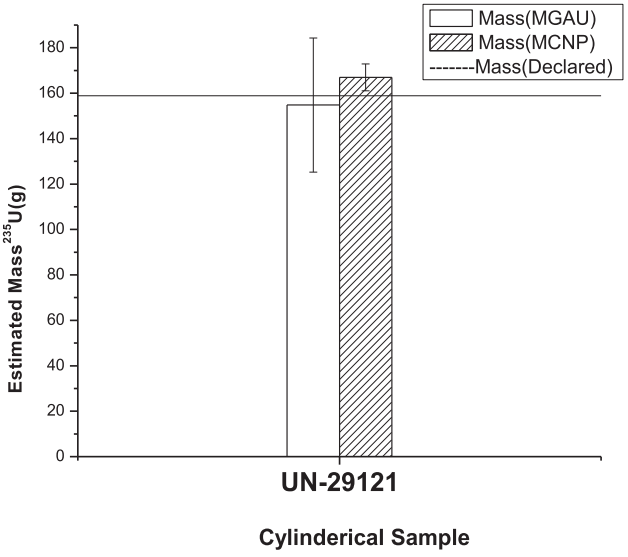


**Fig. 2 – The estimated $^{235}U$ masses based on MCNP and MGUA methods in relation with the declared value for enriched cylindrical sample.**

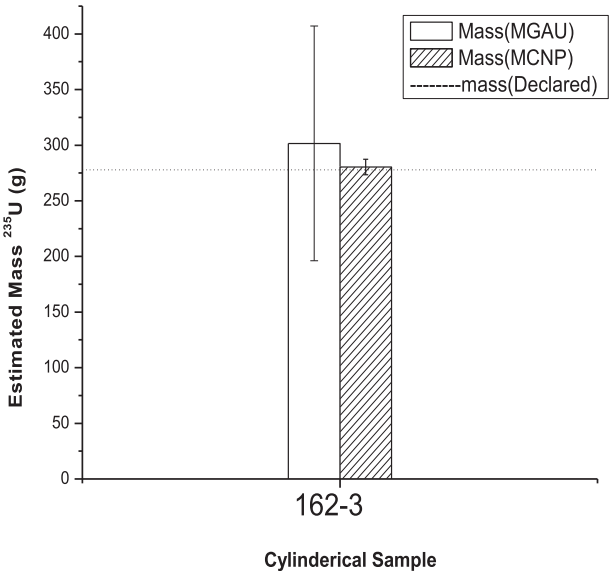


**Fig. 3 – The estimated $^{235}$U masses based on MCNP and MGUA methods in relation with the declared value for natural cylindrical sample.**

element (No. of grams U) were substituted into equation (4) which stated that $E_w$ (wt%) = No. of grams $^{235}$U/No. of grams U × 100. To calculate the mass content (No. of grams) of $^{235}$U-isotope for the two samples at different distances.

Figs. 2–3 and Tables 2–3, show the estimated $^{235}$U mass content by MCNP and MGAU codes in comparison with the declared value for the enriched and natural NM samples, respectively. The relative uncertainties of MGAU and MCNP codes for the enriched and natural samples are 34.99%, 2.46% and 19.08%, 3.52% respectively.

It is observed from the Fig. 2 that, the relative uncertainty of MGAU (34.99%) is relatively high in comparison with the relative uncertainty of MCNP (2.46%) and this is may explained by the large thickness (0.60 cm) of NM shielding which lead to high attenuation for the gamma rays (low count rate for detector) and insufficient measuring time (it is limited as available in a nuclear facility) so the error of MGAU in this case is relatively high. Other factors such as type of shielding material (carbon stainless), the self attenuation for the NM, and low enriched uranium sample further clarify this value taken into account and MGAU software depends mainly on statistics.

It is noticed from Fig. 3 that the relative uncertainty of MGAU (19.088%) is relatively high in comparison to the relative uncertainty of MCNP (3.520%) and that the estimated $^{235}$U mass using MCNP calculations is not in agreement with the declared mass value within the calculated uncertainty. It is expected that some discrepancies were found, the differences could be referred to the deviation of sample location from the designed location and/or the sample non-alignment. In such cases an error of systematic type is expected, both for MCNP calculations and experimental measurements. The sample dimensions are larger in comparison with the dimensions of experimental setup. Such effect was also expected for other samples with relatively large dimensions. The relative uncertainty in the obtained $^{235}$U mass content values calculated by MCNP code were found in comparison with their value which published by the IAEA (International Target Values) (ITVs-2010), the state-of-practice value for $^{235}$U abundance measured using HPGe detector was found to has the value of 3.6% for the relative uncertainty for low enriched uranium oxides (LEU) samples. While the obtained relative uncertainty overall results for the present work was found to have a maximum value of less than 3.6%, which could be accepted in comparison with nearly similar cases of ITVs (STR-368International Target Values 2010 for Measurement Uncertainties in Safeguarding Nuclear Materials Vienna. November, 2010).

It's clear from Tables 2 and 3 that, the relative accuracy value of MCNP is greater than MGAU for the natural uranium sample. Whereas, it is slightly lower than MGAU for enriched sample. Accordingly, the MCNP code may be considered as better approach than the MGAU by using it in a new semi-absolute non-destructive assay technique which has been developed to verify the mass content of $^{235}$U in the large sizes nuclear material samples of different enrichment through combination of experimental measurements and its calculations (version MCNP5) for determination of the $^{235}$U mass content by using the equation (2) which stated that $C_R = M\, S_a\, \varepsilon_a$ where $\varepsilon_a$ is the absolute full energy peak efficiency which calculated from MCNP code and (M) is the mass content of $^{235}$U isotope.

## 6. Conclusion

A new semi- absolute non-destructive assay technique had been developed to verify the $^{235}$U mass content in large sizes nuclear material samples of natural and enriched uranium. It was found that, the relative uncertainties of MGAU and MCNP codes for the enriched and natural samples are 34.99%, 2.46% and 19.08%, 3.52% respectively. The MCNP code seems to be

**Table 2 – The estimated $^{235}$U mass content for enriched and natural in cylindrical containers based on MGAU measurements in comparison with the declared value.**

| Sample ID | | DIST (cm) | U mass (g) ± RSD | | | MGAU (U-enrichment), $E_G$ ±RSD | Relative accuracy (MGAU) $R_{DG}$ % |
|---|---|---|---|---|---|---|---|
| | | | Total mass, $M_T$ | Declared mass, $M_D$ | MGAU mass, $M_G$ | | |
| Enriched | 162-3 | 111.7 | $20.14 \times 10^3$ ±0.005 (%) | 277.93 ±0.004 (%) | 301.59 ±34.99 (%) | 1.49 ± 35.17 (%) | 8.51 |
| Natural | UN-29121 | 96.7 | $22.05 \times 10^3$ ±0.05 (%) | 158.84 ±0.04 (%) | 154.79 ±19.08 (%) | 0.702 ± 19.13 (%) | 2.55 |

**Table 3 – The estimated $^{235}U$ mass content for enriched and natural Uranium in cylindrical containers based on MCNP calculations in comparison with the declared value.**

| Sample ID | | Distance (cm) | U mass (g) ± RSD% | | $C_R(sec^{-1})$ ±RSD | $\varepsilon_{ab} \times 10^{-7}$ ±RSD | Relative accuracy (MCNP)% $R_{DM}$ % |
|---|---|---|---|---|---|---|---|
| | | | Declared mass, $M_D$ | MCNP mass, $M_M$ | | | |
| Enriched | 162-3 | 111.7 | 277.930 ± 0.004 | 280.36 ± 2.46 | 1.56 ± 1.35 (%) | 1.29 ± 2.06 (%) | 0.87 |
| Natural | UN-29121 | 96.7 | 158.840 ± 0.040 | 167.85 ± 3.52 | 1.71 ± 1.01 (%) | 2.37 ± 3.38 (%) | 5.67 |

better than MGAU code by using it in a new semi- absolute non-destructive assay technique through combination of its calculations (version MCNP5) and experimental measurements for determination of the $^{235}U$ mass content in the samples of higher enrichment as mentioned above in equation (2). As a result the importance of the Mont Carlo simulation code as a powerful tool to verify the $^{235}U$ mass content of large sizes NM samples with an experimental method has been clarified and it's recommended to use MCNP simulation code for verification the $^{235}U$ mass content of the large sizes nuclear materials samples.

## REFERENCES


Anilkumar S, Deepa AK, Narayani K, Rekha AK, Achuthan PV, Krichnmachari G, et al. Estimation of $^{235}U$ concentration in some depleted uranium samples by high resolution gamma-ray spectrometry using 185 keV and 1001 keV gamma-energies of $^{235}U$ and 234m Pa. J Radioanal Nucl Chem 2007;274:161.

Ashrfi S, Likar A, Vidmar T. Precise modeling of a coaxial HPGe detector. Nucl Instr Meth 1999;438:421.

Avignone III FT. A Monte Carlo code for predicting the response of a Ge–NaI(T1) Compton suppression spectrometer. Nucl Instr Meth 1980;174:555.

Conlin JL, Tobin SJ. Predicting fissile content of spent nuclear fuel assemblies with the passive neutron albedo reactivity technique and Monte Carlo code emulation. Latin American Section (LAS)/American Nuclear Society (ANS); 2011.

de Castro NV, Levesque RJA. Photopeak and double-escape peak efficiencies of germanium lithium drift detectors. Nucl Instr Meth 1967;46:325.

Debertin K, Grosswendt B. Efficiency calibration of semiconductor detectors by primary standard sources and Monte Carlo calculations. Nucl Instr Meth 1982;203(1–3):343.

El-Gammal W. Verification of 235U mass content in nuclear fuel plates by an absolute method. Nucl Instr Meth 2007;570:446–53.

Ewa IOB, Bodizs D, Czifrus Sz, Molnar Zs. Monte Carlo determination of full energy peak efficiency for a HPGe detector. Appl Radiat Isot 2001;55(1):103.

Fehrenbacher G, Meckbach R, Jacob P. Unfolding the response of a Ge detector used for in-situ gamma-ray Spectrometry. Nucl Instr Meth 1996;454:383.

Hamed AA, Badawi EA, El-Gammal W, Darweesh M. On the absolute measurement of some nuclear material samples. J Nucl Radiat Phys 2010;5:25.

Hurtado S, Garcıa-Leon M, Garcıa-Tenorio R. Monte Carlo simulation of the response of a germanium detector for low-level spectrometry measurements using GEANT4. Appl Radiat Isot 2004;61:139–43.

Kamboj S, Kahn B. Use of Monte Carlo simulation to examine gamma-ray interactions in germanium detectors. Radiat Meas 2003;37(1):1.

Karamanis D. Efficiency simulation of HPGe and Si(Li) detectors in γ- and X-ray spectroscopy. Nucl Instr Meth 2003;505(1–2):282.

Keyser RM. Resolution and sensitivity as a function of energy and incident geometry for germanium detectors. Nucl Instr Meth 2004;213:236.

Kumawat H, Venkata PPK. Development of ICMC-1.0 Monte Carlo code for neutron and Particle transport. BARC News Lett 2013;332:1.

Lepy MC, Altzitzoglou T, Arnold D, Bronson F, Capote Noy R, Décombaz M, et al. Intercomparison of efficiency transfer software for gamma-ray spectrometry. Appl. Radiat. Isot 2001;55:493.

Matussek P. Accurate determination of the $^{235}U$ isotope abundance by gamma spectrometry. KfK 3752. A user's manual for the certified reference material EC-NRM-171/NBS-SRM-969. Institut fur Kernphysik, Kernforschungszentrum Karlsruhe; May 1985.

Meixner C. A Monte Carlo program for the calculation of gamma-ray spectra for germanium detectors. Nucl Instr Meth 1974;119:521.

Michel C, Emling H, Grosse E, Azgui F, Grein H, Wollersheim HJ, et al. Monte Carlo simulation of complex germanium detector systems and Compton suppression spectrometers. Nucl Instr Meth 1986;251(1):119.

Reilly DO, Ensslin NO, Smith Jr HA. Passive Nondestructive Assay of Nuclear Materials. U.S: Nuclear Regulatory Commission, Los Alamos National Laboratory; 1991.

Reilly TD, Martain ER, Parker JL, Speir LG, Walton RB. A continuous in-line monitor for UF6 enrichment. Nucl Technol 1974;23.

Rodenas J, Martinavarro A, Rius V. A validation of the MCNP code for the simulation of Ge-detector calibration. Nucl Instr Meth 2000;450:88–97.

Salgado CM, Conti CC, Becker PHB. Determination of HPGe detector response using MCNP5 for 20– 150 keV X-rays. Appl Radiat Isot 2006;64(6):700.

STR-368 International. Target values 2010 for measurement uncertainties in safeguarding nuclear materials Vienna; Nov. 2010.

Varley BJ, Kitching JE, Leo W, Miskin J, Moore RB, Wunsch R, et al. Investigations of the response of germanium detectors to monoenergetic electron, positron and gamma ray beams. Nucl Instr Meth 1981;543(3):190.

Wainio KM, Knoll GF. Calculated gamma ray response characteristics of semiconductor detectors. Nucl Instr Meth 1966;44(2):213.

Yücel H, Dikmen H. Uranium enrichment measurements using the intensity ratios of self-fluorescence X-rays to 92* keV gamma ray in UXKα spectral region. Talanta 2009;78:410–7.