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Utilization of MCNP Code and ISOCS Software to Develop a Correlation For Comparative Measurements of Uranium Oxide Standards

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Abstract

Comparative studies of absolute efficiency and ^{235}U content in samples using different detectors are useless unless the detector specifications are known. The paper tries to overcome the difficulties facing MCNP code which related to geometry by the aid of ISOCS software. Also, it overcomes the issue of comparing absolute efficiency outcomes from detectors of different geometry using a mathematical model and measured standards. The results of measured count rates, absolute efficiency and ^{235}U mass for five uranium oxide (U_3O_8) standards that have different enrichments were used to perform a correlation between different gamma detectors. The correlation was concluded using three Hyper-Pure Germanium detectors (HPGe) that have different crystal diameters 7.62cm, 8.89cm and 9.4cm. The predicted count rates for the assayed standards were estimated at 25cm away from the three detectors. The developed model was validated by comparison with the certified and calculated ^{235}U mass. A correction for count rate is accomplished to estimate ^{235}U mass accurately by avoiding the small uncertainty that comes from count rate to absolute efficiency ratio. The results are highly accurate with an average accuracy of about 0.13%for D1, 0.09%for D2 and 0.01%for D3.

1. Introduction

During the inspection process, the composition of the supervised material and its mass is a piece of valuable information. Also, when doing quality activities on uranium standards using gamma detectors information like absolute efficiency, count rates and the mass of the isotope U-235 is valuable for comparison and verification purposes. Uranium is a gamma emitter that consists of the well known three isotopes U-235, U-234 and U-238. Gamma rays generated from the disintegration of uranium samples with a certain intensity are a function of the nuclear material mass. Gamma rays corresponding to the energy 185.7 keV are characteristic for estimating ^{235}U enrichment [1-4].

Methods used for assaying nuclear material may need corrections. Corrections depend on many factors like the geometry of the material, its density, and its properties [5–7]. A general-purpose Monte Carlo N-Particle code abbreviated as MCNP can be employed to transport photon, neutron, electron and coupled neutron/photon/electron. Simulating single particles and registration of tallies of its behavior introduce replies with the aid of the Monte Carlo method. MCNP code handles three dimensions configuration of any material in cells that consist of surfaces and can view it [8].

The NMs in this study are Safeguarded and located in (Key Measurement Point (KMP E) of Location Outside Facility (ETZ) at Egyptian Nuclear and Radiological Regulatory Authority (ENRRA). This work includes the use of experimental data outcomes from the measurements of five uranium oxide standards and using it to develop a model to predict the gamma parameters using different detectors. The model is validated using the manufacturing data for the assayed uranium oxide standards.

2. Methodology

Gamma spectrometers are fitted with detectors necessary for photon energy measurement. Through doing a correlation of the photo-peaks to the characteristic energies of each isotope, a spectrum can thus be utilized to distinguish the gamma-emitting nuclide in a substance. Also, to determine the relative occurrence of isotopes, the correlation of various peak intensities can be used. ^{235}U mass determination is the ultimate objective of the non-destructive assay technique. To measure the count rate, a standard nuclear material (SNM) is opposite to the detector at distance ‘‘D’’. This could be given as, [9]

$$C_R = M_5 S_{a5}\, \boldsymbol{\varepsilon_a} \ldots\ldots\ldots\ldots (1)$$

where C_R is the net count rate for the SNM at distance D, M_5 is the mass of the ^{235}U in SNM, S_{a5} is the specific activity of the 185.7 keV energy line, and $\boldsymbol{\varepsilon_a}$ represents the absolute efficiency of the detector for SNM at the 185.7 keV energy line. The absolute efficiency could be calculated using MCNP-5 where the created input file depends on the SNM certificate and the dimensions details of the used detector [10]. From the first equation, the count rate and absolute efficiency for two detectors measuring the same sample at the same conditions can be represented by the following equation:

$$(C_R/\boldsymbol{\varepsilon_a})1 = (C_R/\boldsymbol{\varepsilon_a})2 \ldots\ldots\ldots\ldots (2)$$

C_{R1} is the experimental net count rate for the SNM at distance D, $\boldsymbol{\varepsilon_{a1}}$ represents the absolute efficiency of the detector for SNM at the 185.7 keV energy line measured by MCNP5. C_{R2} is the predicted experimental net count rate for the SNM at distance D, $\boldsymbol{\varepsilon_{a2}}$ represents the ISOCS absolute efficiency of the detector for SNM at the 185.7 keV energy line measured by MCNP5.The second equation suppose the uranium in the assayed sample has the same chemical formula also the mass of uranium and the geometry of the sample is the same. It is valid when comparing samples that have the same previous conditions at certain enrichment. The need for such an equation is necessary when detectors are of unknown geometry.

3. Experimental setup and techniques

3.1. Measurements

A group of standard samples in a cylindrical form was utilized to execute the measurements. The NMs consist of five NBS-SRM-969 with different ^{235}U content were in a cylindrical aluminum can containing 200.1 g of U_3O_8 [**11**]. The external radius of the can is 40 mm and the internal one is 35 mm, the height is 89 mm and the fill height of the compact powder for all samples is (20.8±0.5) mm except for SRM969-446 sample whose fill height is (15.8±0.5) mm.

The sample was centered in front of the detector; Figure 1 shows the configuration setup to count the pulses at 185.7 keV energy line. Before measuring the samples energy calibration was done and the dead time was optimized to be low by using appropriate distance. For all measurements, the distance from the sample to the detector AL cap was at an optimum distance of 25 cm at which dead time for the detector is less than 1%. The measuring times were 2400 sec per measuring cycle where the measurements were obtained for three runs each.

These samples with the above mentioned set up and conditions were measured in the work done by Sameh E. Shaban et al (2019) [9]. The same samples with all the above conditions were modeled using ISOCS software and simulation was done to get the absolute efficiency for three types of gamma detectors. Figure (2) shows the set up of standard uranium oxide at a distance of about 25cm from the detector. Information concerning the used detectors is listed in table 1.

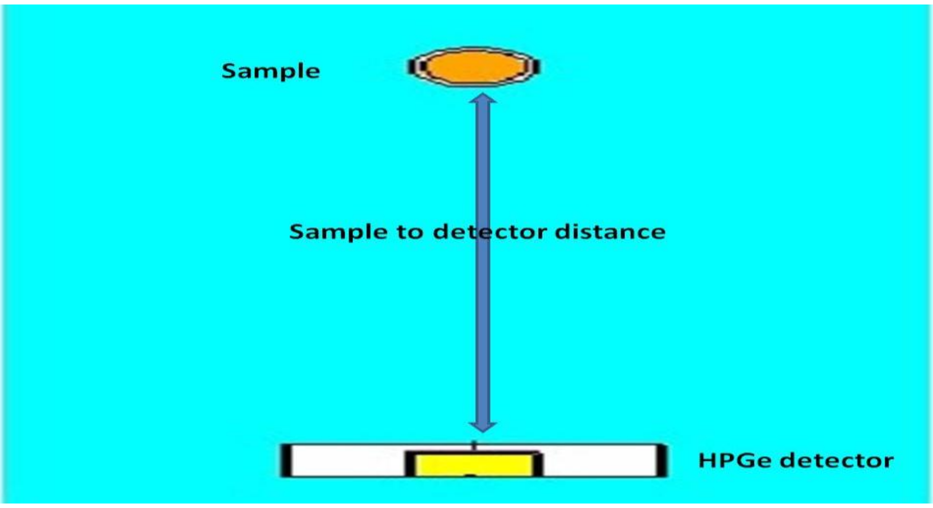

Figure 1: Detector and sample set up in MCNP5

Table 1: Specifications of the gamma detectors.

Detector ID	First detector (D1)	Second detector (D2)	Third detector (D3)
Type	HPGe	HPGe	HPGe
Diameter (cm)	7.62	8.89	9.4

The results obtained from ISOCS software were compared with the experimental count rates and measured absolute efficiency from MCNP5 using the second equation. Once the correlation is performed the predicted count rate outcomes. The mass of U-235 was calculated and compared with that obtained from the MCNP5 to validate that correlation.

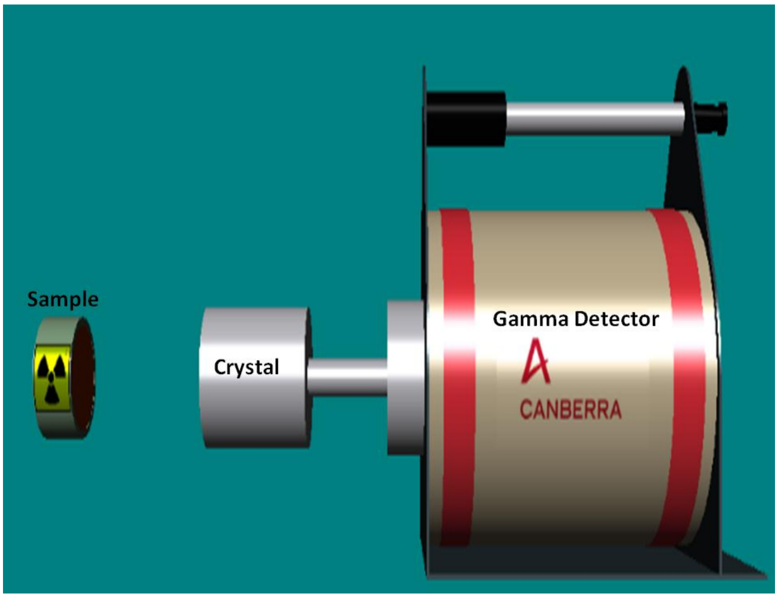

Figure 2: Detector and sample set up in ISOCS

3.2. Monte Carlo Modeling for Measuring System

Simulation using Monte Carlo is random numbers that happen during the simulation in a sequence manner. When this sequence of random numbers is repeated, the simulation will lead to results that match with those obtained from the first sequence with some 'statistical error' [12]. The General Monte Carlo Code (MCNP-5) was utilized to get information concerning the absolute efficiency of the detector [13-16]. The geometry specifications of the planar HPGe detector and SNM were described as many details as possible in order to build and simulate the model correctly. The histories used in the prepared input files were 10^8 histories with run time 30 minutes. The specifications of the used laptop are 2.5 GHz Intel Core i7 processor since a tally F8 is used to determine the pulse height of the detector. The absolute efficiency of the detector at 185.7 keV energy line was calculated by means of that tally.

3.3. ISOCS Modeling for Measuring System

ISOCS™ software makes it possible to get the absolute efficiency curve for certain energy range by simulation using known or guessed geometry and chemical form of measured item. It supplies various templates for geometry that cover wide range of possible samples shapes (cylinders, pipes, boxes as well as more complex geometries). This method avoids wasting time in calibration measurements. The basic geometry templates included with the ISOCS calibration software was used to generate the efficiency file [17]. After entering the parameters, a preliminary check of the geometry validity is done the efficiency file then used for, in exactly the same manner as those produced by traditional "calibrated source" calibrations, the efficiency calibration process. Total U_T, ^{238}U and ^{235}U mass contents estimated also based on the ISOCS™ software calibration [18].

4. Results and Discussion

The ratio between count rate (CR) for energy line 185.7 keV and absolute efficiency at different enrichment values of SNM are given in table 2. The results in table 2 was obtained using HPGe detector with model Canberra GL0515R. It is clear that when sample enrichment increased, the ratio between sample count rate to absolute efficiency increases. Sample U1 which has enrichment value 0.31% has the lowest value of the previous ratio. Samples U2,U3 and U4 have enrichment values 0.71%, 1.94 % and 2.95% respectively and the ratio increase by increasing the value of enrichment. Sample U5 that is low enriched (4.46%) has the highest ratio of count rate to absolute efficiency.

Table 2: The count rate to absolute efficiency ratio for U_3O_8 standards.

Sample ID	Count rate to absolute efficiency ratio
U1	24474.49053
U2	56625.07035
U3	155851.6674
U4	234346.025
U5	355275.1411

The ratio in table 2 was used to obtain the predicted count rate through two steps. The first step is the calculation of absolute efficiency for the required detector. The second step involves predicting the value of count rate. In table 3 the absolute efficiency was measured for the first detector. It is obvious that the count rate increases by increasing the value of enrichment also the same trend is remarkable in the values of absolute efficiency.

Tables 4 & 5 show the predicted values of count rates and the values of absolute efficiencies. It is clear the same trend of relation between enrichment value and count rate that presents in table 3 is also observed for the other two detectors.

Table 3: Absolute efficiency and predicted count rate for (D1) detector.

Sample ID	Predicted count rate for D1 $C_R \pm (\sigma_{CR})$ (s^{-1})	Absolute efficiency $\varepsilon_{ab} \pm \sigma_{\varepsilon ab}$
U1	6.0930710±0.2752525	0.0002490±0.0000199
U2	14.1768790±0.6305688	0.0002500±0.0000200
U3	39.1307691±1.7226826	0.0002511±0.0000201
U4	58.6743860±2.6190846	0.0002500±0.0000200
U5	116.4602570±5.2234200	0.0003280±0.0000262

Table 4: Absolute efficiency and predicted count rate for (D2) detector.

Sample ID	Predicted count rate for D2 $C_R \pm (\sigma_{CR})$ (s^{-1})	Absolute efficiency $\varepsilon_{ab} \pm \sigma_{\varepsilon ab}$
U1	9.3978130±0.4271255	0.0003840±0.0000307
U2	21.8663370±0.9784907	0.0003860±0.0000308
U3	60.3548050±2.6731879	0.0003873±0.0000309
U4	90.4997480±4.0641875	0.0003860±0.0000308
U5	179.6395460±8.0548341	0.0005060±0.0000405

Table 5: Absolute efficiency and predicted count rate for (D3) detector.

Sample ID	Predicted count rate for D3 $C_R \pm (\sigma_{CR})$ (s^{-1})	Absolute efficiency $\varepsilon_{ab} \pm \sigma_{\varepsilon ab}$
U1	6.4573250±0.2932338	0.0002640±0.0000211
U2	15.0243870±0.6717618	0.0002650±0.0000212
U3	41.4699468±1.8352196	0.0002661±0.0000213
U4	62.2027000±2.7901805	0.0002650±0.0000212
U5	123.4222290±5.5237696	0.0003470±0.0000278

The results obtained from tables 3,4 and 5 for count rate and absolute efficiency showed that the used detectors have different geometries. It also showed some geometrical similarities between the first and the third detector.

The validation of the used procedure for predicting count rate using ratio method indicates that the mass of U-235 is the same for the three detectors (0.530513, 1.227414, 3.3782668, 5.079724 and 7.701003 gm). These values are accurate when compared with the manufacturer but there is a problem. The problem is the values logically should be different so a correction must be done. Table 6 shows the mass values of U-235 after correction for count rate using the known value of mass. It is clear that the mass values increases by increasing the enrichment value also, the mass value differs from detector to another. The results show that the model is valid.

Table 6: The results of U-235 mass for all detectors.

Sample ID	Manufacturer U-235 mass (g)	U-235 mass by D1 (g) M (g) $\pm\sigma_M$	U-235 mass by D2 (g) M (g) $\pm\sigma_M$	U-235 mass by D3 (g) M (g) $\pm\sigma_M$
U1	0.526	0.525892±0.023962	0.526121±0.024111	0.525804±0.024111
U2	1.205	1.203443±0.054673	1.206851±0.054948	1.209083±0.054948
U3	3.292	3.2892777±0.14877	3.2912390±0.149727	3.2911057±0.149551
U4	5.005	5.004024±0.227087	5.006769±0.228228	5.006006±0.228228
U5	7.567	7.538317±0.345055	7.587848±0.346451	7.549867±0.346412

Conclusion

MCNP code is applied to measure the absolute efficiency of HPGe detector that has known geometry and crystal diameter 8cm. The count rate in this case was measured experimentally for five set of uranium standards that have different enrichments. Detectors that have different geometry and crystal diameter 7.62cm, 8.89cm and 9.4cm were modeled using ISOCS software for the same standards to predict the experimental count rates. ISOCS was used because the specifications of the detectors were not known so there is no mean to model it by MCNP. The proposed equation that depends on the relation between count rate and absolute efficiency solves the problem. By this equation, the analyst can predict the count rates that will appear experimentally for unattended detectors. The validity of the equation was tested by measuring the mass of U-235 and the results were accurate. It is recommended to correct for count rate to facilitate the comparative measurements between different detectors.

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