Feasibility Study on the U Factor Analysis of UO_2 Pellets using Gamma Spectroscopy

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1. Introduction

The International Atomic Energy Agency (IAEA) defines nuclear safeguards as "the timely detection of diversion of nuclear material from peaceful nuclear activities to the manufacture of nuclear weapons or of other nuclear explosive devices…" [1]. Special nuclear material (SNM) is defined as the material subjected to IAEA safeguards. The ROK, as a member state of IAEA, is obligated to control domestic SNMs based on state system of accounting and control (SSAC) [2]. The Korea Institute of Nuclear non-proliferation and Control (KINAC) is committed to the control of SNM in the ROK by the Nuclear Safety and Security Council (NSSC). KINAC has to perform independent verification on the SNM information declared by domestic license holders due to the article 4 of NSSC notification (No. 2017-83) [3].

Since the direct verification of all nuclear materials in a facility is almost impossible, IAEA verifies the amount of SNM based on sampling. The conventional IAEA sampling method considers three levels of verification process (gross, partial and bias defect verification). The corresponding sample sizes for each defect level are then calculated. The characteristics and purpose of each defect verification are summarized in Table 1 [4].

Table 1. Characteristics of different defect types.

Type of defects	Target	Location of verification	Methods
Gross defect	Material type (NU, EU,…)	On-site	Gamma spectroscopy (NDA)
Partial defect	Amount of SNM (^{235}U)	On-site	Weighing, Gamma spectroscopy (NDA)
Bias defect	Amount of SNM (^{235}U)	Analysis laboratory	Chemical analysis (DA)

※ NDA: Non-destructive assay, DA: Destructive assay

IAEA applies operator declared U factor for partial defect verification due to the absence of an NDA based U factor analysis method. However, the domestic notification requires to verify operator declared U factor and SNM quantity simultaneously. As a result, a novel "NDA based U factor analysis method" is required to apply IAEA's sampling method on national inspection.

The purpose of this study is to demonstrate the feasibility of analyzing the U factor of bulk UO_2 pellets using the gamma spectrum. The suggested method does not require additional burden for both inspectors and operators, since gamma spectroscopy is already applied for on-site gross and partial defect verifications. The results of this study can be a basis of applying the IAEA's sampling method to KINAC's national inspection under the NSSC's notification on the accounting of SNM.

2. Methods and Results

2.1 Methods

This study made the following assumptions to simplify the problem:

1) Daughter nuclides of ^{235}U and ^{238}U are separated during fuel fabrication process
2) Enrichment of a target UO_2 pellet are known using the enrichment meter method (intensity of 185.7 keV (^{235}U) is given)
3) Reference pellets with same geometry but different enrichment and U factor exist
4) Detector's energy response function exists

UO_2 pellets in fuel fabrication plants (FFPs) consist of uranium isotopes (^{234}U, ^{235}U, ^{238}U), daughter nuclides of uranium, oxygen, and burnable poison (Gd, Er). Since the U factor of a pellet is affected by the concentration of burnable poisons, it can be calculated by measuring the intensity of uranium's characteristic X ray generated by internal gamma rays.

The energy range of uranium's characteristic X rays are around 90 keV (K_α) and 110 keV (K_β), which are overlapped with gamma peaks from a pellet. Therefore, the net intensity of uranium's characteristic X ray can be calculated by subtracting the intensity of gamma peaks from entire counts between 80 and 120 keV.

According to the 1st assumption, major radioisotopes with gamma emission in a pellet are ^{234}U, ^{235}U, ^{238}U, ^{231}Pa, ^{234m}Pa, ^{230}Th, ^{231}Th, and ^{234}Th. All nuclides, except ^{234}U and ^{230}Th, satisfy secular equilibrium with ^{235}U and ^{238}U. The count rate of a gamma peak can be calculated using equation (1). According to the 2nd and 4th assumptions, count rate of gamma peaks from ^{235}U series and ^{238}U series are calculated using equation (2) and (3) respectively. Since the daughter nuclides of ^{235}U and ^{238}U are at secular equilibrium, their activity are equal to the activity of ^{235}U and ^{238}U. Therefore, net count rate of uranium's characteristic X ray is calculated using equation (4).

$$C = \lambda_X w_X N_U Y(E_\gamma)\varepsilon(E_\gamma)\varepsilon_{etc} \quad (1)$$

$$\mathrm{C}(\mathrm{E}_\gamma, {}^{235}U) = \mathrm{C}(185\ \mathrm{keV})\frac{Y(E_\gamma)\varepsilon(E_\gamma)}{Y(185\ keV)\varepsilon(185\ keV)} \quad (2)$$

$$\mathrm{C}(\mathrm{E}_\gamma, {}^{238}U) = \mathrm{C}(185\ \mathrm{keV})\frac{\lambda_{238}(1-w_{235})Y(E_\gamma)\varepsilon(E_\gamma)}{\lambda_{235}w_{235}Y(185\ keV)\varepsilon(185\ keV)} \quad (3)$$

$$\mathrm{C}(\mathrm{X_U}) = \sum_i\ \mathrm{C(i)} - \left(\sum_j \mathrm{C}(\mathrm{E}_{\gamma,\mathrm{j}}, {}^{235}U) + \sum_k \mathrm{C}(\mathrm{E}_{\gamma,\mathrm{k}}, {}^{238}U)\right) \quad (4)$$

where,
C: Net count rate of a gamma peak,
$C(E_\gamma, {}^{235}U)$: Net count rate of a gamma peak from ^{235}U
$C(E_\gamma, {}^{238}U)$: Net count rate of a gamma peak from ^{238}U,
$C(X_U)$: Net count rate of uranium's characteristic X ray between 80 and 120 keV,
$C(i)$: Net count rate of channel i between 80 and 120 keV,
j: j th gamma peak from ^{235}U between 80 and 120 keV,
k: k th gamma peak from ^{238}U between 80 and 120 keV,
λ_X: Decay constant of uranium isotope X (s^{-1}),
w_X: Enrichment of uranium isotope X (at%),
N_U: Number of uranium atoms in a pellet,
$Y(E_\gamma)$: Yield of gamma($E = E_\gamma$) emission,
$\varepsilon(E_\gamma)$: Detector's energy efficiency at ($E = E_\gamma$),
ε_{etc}: Other detector efficiencies.

2.2 Results for benchmark cases

This study verified the feasibility of the "gamma spectroscopy based U factor analysis" using the MCNPX code, due to the limited accessibility on reference pellets with different U factors (Gd_2O_3 poison concentration) and ^{235}U enrichments. A simplified detector geometry was applied for simulation, as depicted in figure 1.

This study simulated the measurement results of 32 reference UO_2 pellets with four different U factors (Pure UO_2, 4wt% Gd_2O_3, 6wt% Gd_2O_3, 8wt% Gd_2O_3) and eight different enrichments (0.72, 1.5, 2.0, 2.5, 3.0, 3.5, 4.0, 4.5 wt%) using two types of gamma detectors (NaI(Tl) and HPGe).

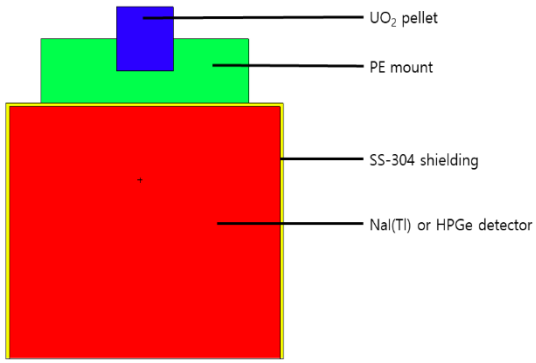


Fig. 1. Simplified detector geometry

Gamma source in a UO_2 pellet was calculated using the OrigenArp code in SCALE 6.1 package [5] and gamma information of KAERI's nuclear database [6].

The OrigenArp code calculates the relative mass of gamma emitting radioisotopes (^{234}U, ^{235}U, ^{238}U, ^{231}Pa, ^{234m}Pa, ^{230}Th, ^{231}Th, and ^{234}Th) in pure UO_2 at 1 year after its manufacture. The half-life of each radioisotopes and relative yield of all gamma peaks were then applied to the OrigenArp results. The results were normalized to become the source of MCNPX input files. This research neglected gamma peaks whose intensity is smaller than 10^{-4} times of total gamma intensity. Figure 2 depicts the relative gamma source distribution of a pure UO_2 pellet with 4.5wt% ^{235}U enrichment.

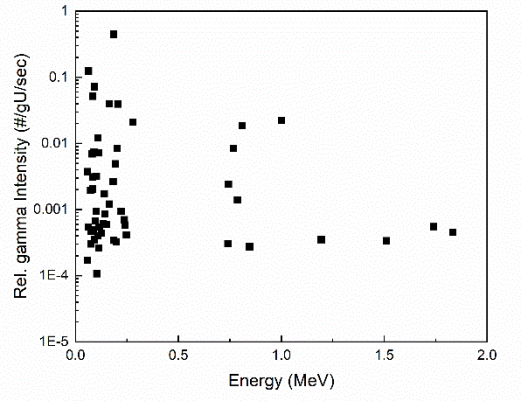
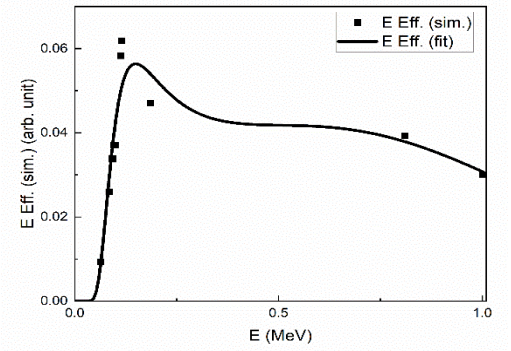
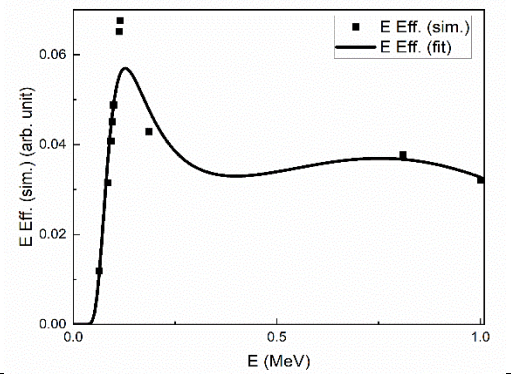


Fig. 2. Gamma source distribution in a pellet

This study also simulated the energy efficiency of NaI(Tl) and HPGe detectors with point energy sources between 60 keV and 1,001 keV. The energy efficiency was then fitted to equation (5). Energy efficiency curve of two detector types and constants of equation (5) are depicted in Table 2.

Table 2. Energy efficiency curves for NaI(Tl) and HPGe

NaI(Tl)		
Constants)	a: -19.044,	b: -175.89,
c: -605.62,	d: -918.45,	e: -520.06.
		
HPGe		
Constants)	a: -0.4585,	b: -1.9927,
c: -2.5066,	d: -0.9840,	e: -3.4179.
		

$$ln(\varepsilon(E)) = a(ln(E))^4 + b(ln(E))^3 + c(ln(E))^2 + d(ln(E)) + e \quad (5)$$

where,
$\varepsilon(E)$: Energy efficiency of a detector,
E: Energy of a gamma photon (MeV),
a, b, c, d, e: Constants of equation (5).

This study then simulated gamma and X ray spectrum of the 32 reference UO_2 pellets using the MCNPX code. Pulse height tally (F8 tally) was selected to simulate the net count rate of each peak. Gaussian energy broadening was applied for both NaI(Tl) and HPGe [7]. The width of individual energy bin was 0.001 MeV. Number of particles for simulations were 5×10^8 and 5×10^7 for NaI(Tl) and HPGe respectively. The number of simulated particles for HPGe is smaller than NaI(Tl) due to its higher energy resolution. The background of simulated spectrums was calculated using the SNIP method [8]. The net count rate of a gamma peak was then calculated by subtracting the background count rate from total count rate. Figure 3 depicts the simulated spectrums of 4.5 wt% enriched pure UO_2 using NaI(Tl) and HPGe.

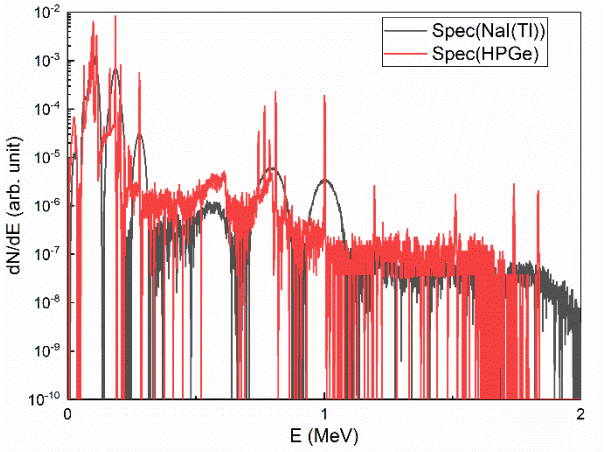


Fig. 3. Gamma spectrum of a pure UO2 pellet (4.5 wt% ^{235}U).

Net count rates of characteristic X ray were calculated using equations (2), (3), and (4), for all cases. Table 3 (NaI(Tl)) and Table 4 (HPGe) describes the results of simulated the net count rates per gU-second. Results in Table 3 and 4 indicate the net count rate of uranium's characteristic X ray is affected by ^{235}U enrichment as well as U factor.

As U factor increases, the intensity of gamma photons inside a pellet and probability of generating uranium's characteristic X ray increases simultaneously. Due to the reason, net count rate of uranium's X rays and U factor of UO_2 pellets have the 2nd order polynomial relationship, as depicted in equation (6).

As ^{235}U enrichment increases, the intensity of gamma photons with energy higher than uranium's characteristic X ray increases. Therefore, net count rate of uranium's characteristic X ray increases linearly as ^{235}U enrichment increases, as depicted in equation (7).

$$C(X) = A \cdot f_U^2 + B \cdot f_U + C \quad (6)$$
$$C(X) = D \cdot f_w + E \quad (7)$$

where,
$C(X)$: Net count rate of uranium's characteristic X ray,
f_U: U factor of a pellet,
f_w: ^{235}U enrichment of a pellet,
A, B, C, D, E: Constants for equation (6) and (7).

Table 3. Net count rate of uranium's characteristic X rays for all pellet cases using NaI(Tl) detector.

Net X counts (#/gU/sec)		U235 enrichment(wt%)							
		0.72	1.5	2.0	2.5	3.0	3.5	4.0	4.5
Gd_2O_3 (wt%)	0	47	84	107	130	154	176	199	222
	4	46	81	103	126	148	170	193	215
	6	45	80	103	124	145	168	190	212
	8	44	79	101	122	144	166	188	209

Table 4. Net count rate of uranium's characteristic X rays for all pellet cases using HPGe detector.

Net X counts (#/gU/sec)		U235 enrichment(wt%)							
		0.72	1.5	2.0	2.5	3.0	3.5	4.0	4.5
Gd_2O_3 (wt%)	0	9.9	19.4	25.3	31.4	38.6	43	48.6	54.3
	4	9.7	18.9	24.7	30.6	36.5	42	47.6	53.1
	6	9.5	18.8	24.6	30.4	36.0	41.7	47.2	52.7
	8	9.4	18.7	24.6	30.1	35.9	41.3	46.8	52.3

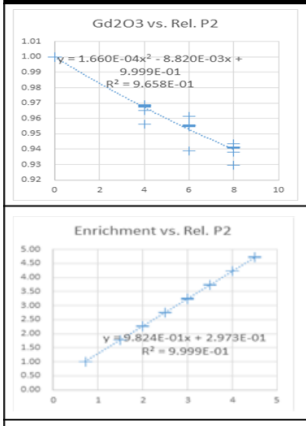
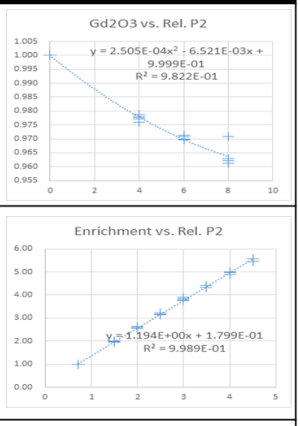
This study calculated the relative net X count rates for the same Gd_2O_3 concentration and ^{235}U enrichment to estimate the constants in equation (6) and (7). Estimated constants (A~E) for NaI(Tl) and HPGe detectors are described in Table 5.

Once net X ray count rate of a reference pellet and the enrichment of a target pellet are given, Gd_2O_3 concentration of the target pellet is calculated using the measured net X ray count rate of a target pellet and equation (8). U factor of the target pellet can be calculated using equation (9).

$$X(f_w, Gd_w) = X(f_{ref}, Gd_{ref}) \frac{A \cdot Gd_w^2 + B \cdot Gd_w + C}{A \cdot Gd_{ref}^2 + B \cdot Gd_{ref} + C} \frac{D \cdot f_w + E}{D \cdot f_{ref} + E} \quad (8)$$
$$f_U = \left(\frac{100 - Gd}{100}\right)\left(\frac{M(U)}{M(UO_2)}\right) \quad (9)$$

where,
$X(f_w, Gd)$: Net X ray count rate for a pellet,
$f_{ref/w}$: ^{235}U Enrichment of reference/target pellets (wt%),
$Gd_{ref/w}$: Gd_2O_3 concentration of reference/target pellets (wt%),
f_U: U factor of target pellets,
$M(UO_2), M(U)$: Molar mass of UO_2 and U.

Table 5. Estimated constants (A~E) for NaI(Tl) and HPGe.

NaI(Tl)	HPGe
	
A: 1.660E-04, B: -8.820E-03, C: 9.999E-01, D: 9.824E-01, E: 2.973E-01	A: 2.505E-04, B: -6.521E-03, C: 9.999E-01, D: 1.194E+00, E: 1.799E-01

3. Discussions

Results of this study indicate the net count rate of uranium's characteristic X ray depends on the ^{235}U enrichment and U factor of an UO_2 pellet. The relative difference of net X ray count rates between a pellet with 6 wt% Gd_2O_3 and a pellet with 8 wt% Gd_2O_3 impurity is 1.5% for NaI(Tl) and 0.69% for HPGe detector. Since the standard uncertainty of a gamma peak follows equation (10), counts required to distinguish those two pellets with 95% confidence interval are 1.685×10^4 for NaI(Tl) and 8.069×10^4 for HPGe.

$$\sigma_{Rel.} = \frac{1}{\sqrt{N}} \quad (10)$$

where,
$\sigma_{Rel.}$: Relative standard uncertainty of gamma count (rate),
N: Net X, gamma ray count (rate).

However, on-site measurement time is limited for safeguards inspection. HPGe is not desirable for national inspection due to its low detection efficiency. Therefore, gamma detectors with high detection efficiency, such as NaI(Tl), are desirable for on-site U factor analysis in national inspection.

4. Conclusions

Results of this study demonstrated the feasibility of U factor analysis using gamma spectroscopy. Once the measurement results of reference pellets and the enrichment of target pellets are given, the suggested method can calculate the U factor of target UO_2 pellets.

Since gamma spectroscopy is already applied to conventional IAEA and national inspection to measure the enrichment of target pellets, the method can be a solution for a NDA based U factor analysis without additional burden. Due to the limited measurement time, gamma detectors with high detection efficiency, such as NaI(Tl), are desirable.

Future works include validation of the simulation results and the accuracy of the method. Validation of simulation will compare calculated results (Monte Carlo simulation) and measured results (gamma spectroscopy). Validation of the accuracy will compare the U factor analysis results using the gamma spectroscopy based method and conventional thermogravimetric method.

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