A Study on the Uranium Enrichment Determination using the Standard-less Gamma Spectrometry Technique

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1. Introduction

Determining uranium enrichment is one of most important characteristics of nuclear materials, and have to be assessed rapidly and accurately to make decisions whether it is a threat to national security as well as it is the unauthorized movement. Current national radiation monitoring regime for the import products has been conducted through radiation portal monitor and dosimetry by the act on the protective action guidelines against radiation on the natural environments. Then the secondary inspections are made by hand-held radioisotopes identification devices. If it is assessed that there exist any suspicious radiological threats, anyone who acknowledge it have to notify the fact as soon as possible to the Nuclear Safety and Security Commission(NSSC) or Korean National Policy Agency(KNPA) by the act on the counter-terrorism for the protection of the citizens and public security. Then national radiological terror response system will be activated, several technical support teams from Korea Institute of Nuclear Safety(KINS), Korea Institute of Nuclear Nonproliferation and Control(KINAC) and Korea Institute of Radiological & Medical Sciences(KIRAMS). If the threat is closely related to nuclear material (i.e. enriched uranium compounds, any kinds of plutonium), KINAC can provide major technical support through preliminary characterization using gamma and X-ray fluorescence spectrometry. Especially, it is well known that general kind of equipment used in first/second inspection are not possible to determine ratio of the isotopes (i.e. the enrichment, type of Uranium). For the alternative to this problem, there is standard-less gamma spectrometry method which is called peak-ratio techniques [1]. For using this technique, it needs to use the high purity Ge detector(HPGe) which have high resolution.

This paper describes that the results of the uranium enrichment determination using HPGe and standard-less technique with commercially utilizing spectrum analysis code. The spectra utilized in this study are used as the same which European Commission-Joint Research Center(EC-JRC) were conducted in their experiments [2].

2. Material and Equipment

The ORTEC Micro Detective, electrically cooled spectrometer, was used for collecting the spectra. The sensor is made of high purity germanium crystal, and have 50mm diameter and 30 mm depth. It has 2.0keV at 1332keV and below 1.0keV at 122keV. The conversion gain from channel to energy is set to 0.36621keV/ch, and the total MCA channel is used at 8192.

CBNM Uranium set known as EC NRM-171 or NBS SRM-969 [3] were used for the experiment. This set contains 200grams of U3O8 compounds in a sealed aluminum can. It also has 2mm Al window and 1mm blank space at the bottom of the container. It consists of 5 type items according to U-235 enrichment as shown in the Table I.

Table I. CRM's Certified Values in Enrichment

Name	U-235(Mass%)	Standard Deviation
CBNM031	0.3163	0.0002
CBNM 071	0.7116	0.0002
CBNM 194	1.9414	0.0005
CBNM 295	2.9477	0.0012
CBNM 446	4.4617	0.0015

3. Analysis Method

Methodologies for analyzing the isotopes composition using emitted radiation from Uranium and Plutonium samples has been well established since few decades in U.S. and Europe [4,5]. Among them, most favorable and well-known one is enrichment meter method. However, this methodology has many limits for utilizing under in-situ conditions. For example, the method strongly depends on sample's intrinsic characteristics such as composition and density, as well as their container material. It needs several data for calibration and corrections prior to enrichment calculation.

An alternative method of these limits is to use ratios among the peaks from each isotopes using the characteristic to emit multi-peaks for Uranium and Plutonium. Representative codes using this method are MGAU(Multi Group Analysis for Uranium) and

FRAM(Fixed energy, Response functions Analysis with Multiple efficiency). This method is not affected by their sample composition and shielding material rarely. Namely, it is possible for unknown sample to calculate their isotope ratios using the equipment not to have calibration and correction aids.

FRAM code 4.4 version distributed by Canberra is used. This code calculates not only Plutonium isotopes ratio but also Uranium enrichment. But, users should be aware of the parameter set in advance which contains details for calculation of isotopes ratios. The code distributers have been developed their parameter sets based on know-how from the U.S. National Laboratories. Those are included as the built-in within the code. Table II shows that the parameter sets to use Uranium analysis.

Table II. Parameter Set List built in FRAM Code

Name	Descriptions
U100keVLEU	Planar, E<=10%, 100keV
U100keVHEU	Planar, E>=2%, 100keV
ULow121_1001Cx	Coaxial, E<70%, 121-1001keV
UHi121_1001Cx	Coaxial, E>10%, 121-1001keV
ULowENR	Coaxial, E<=50%, 1-1024keV
UHiENR	Coaxial, E>20%, 1-1024keV

ULow121_1001Cx parameter was used with some modifications. The parameter is focus on the low enrichment of Uranium and it uses the energy peaks from 121 keV to 1001 keV, and the coaxial type of detector would be used.

4. Results

It assumes that the electrically cooled HPGe detector and FRAM code are used for preliminary characterization in-situ environment of the suspicious radiological terror scenarios. The source to detector distances are 2 and 10 cm, counting times are 5 minutes and 16 hours. These measuring conditions are categorized into 4 Cases as below.

- Case 1 : 10cm and 16 hours
- Case 2 : 2cm and 16 hours
- Case 3 : 10cm and 5 minutes
- Case 4 : 2cm and 5 minutes

Case 2 is assumed to have a best measuring condition and Case 3 is a worst measuring condition because of the distances between the source to detector, and counting time. Unfortunately, Case 1 and 2 of CBNM194 were not used due to no data in the EC-JRC experiment.

4.1. Calculated U-235 Values

Entirely, Fig.1 shows all cases estimate consistently compared to their reference values. CBNM031 is underestimated and CBNM295 is over estimated in Case 3. CBNM295 is underestimated and CBNM446 is over estimated in Case 4.

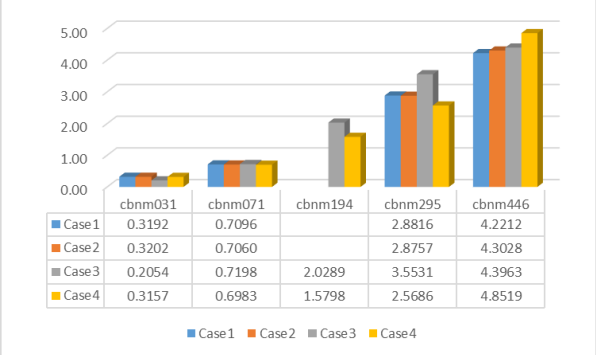

Fig. 1. U-235 Enrichment Values Calculated by FRAM code

4.2. Relative Deviation

Fig.2 shows the relative deviations between reference value and calculated value. As expected in statistics, Case 2 has most correct results except CBNM071. Case 3 is to have entirely largest error except CNMB 174 and 446.

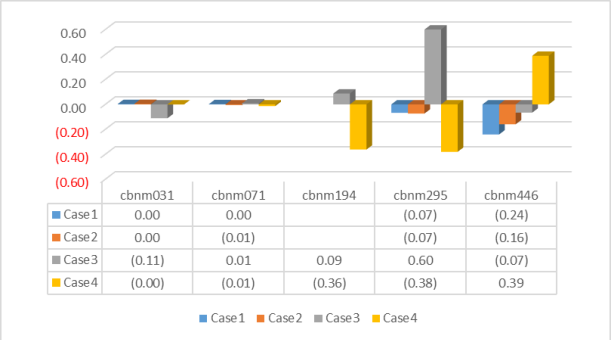

Fig. 2. Relative Deviation of the Calculated U-235 Values by Reference Values

5. Conclusion

This paper describes the evaluation results of the uranium enrichment using standard-less gamma spectrometry technique for determining isotope ratios. Samples are the CBNM Uranium Set which have 5 type of Uranium enrichment. Calculation conditions are divided into 4 cases by the source to detector distance and measuring time. The results show that the calculated enrichments are within the maximum 0.6 % in relative deviation. It is well observed that consistency between calculated and reference largely depends on the distance and measuring time. It means that Case 2 has the most correct results due to short

distance and long measuring time. Even though the results of this study, Uranium enrichment is well estimated in Case 3 within maximum 0.6%. Therefore, this standard-less gamma spectrometry technique is appropriate for determining Uranium enrichment in the contingency situation such as suspicious radiological terror.

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