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## **Original Article**

# Basic characterization of uranium by high-resolution gamma spectroscopy



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## ABSTRACT

A basic characterization of uranium samples was performed using gamma- and X-ray spectroscopy. The studied uranium samples were eight types of certified reference materials with  $^{235}$ U enrichments in the range of 1–97%, and the measurements were performed over 24 h using a high-resolution and high-purity planar germanium detector. A general peak analysis of the spectrum and the XK $_{\alpha}$  region of the uranium spectra was carried out by using HyperGam and HyperGam-U, respectively. The standard reference sources were used to calibrate the spectroscopy system. To obtain the absolute detection efficiency, an effective solid angle code, EXVol, was run for each sample. Hence, the peak activities and isotopic activities were determined, and then, the total U content and  $^{234}$ U,  $^{235}$ U, and  $^{238}$ U isotopic contents were determined and compared with those of the certified reference values. A new method to determine the model age based on the ratio of the activities of  $^{223}$ Ra and  $^{235}$ U in the sample was studied, and the model age was compared with the known true age. In summary, the present study developed a method for basic characterization of uranium samples by nondestructive gamma-ray spectrometry in 24 h and to obtain information on the sample age.

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#### 1. Introduction

As a prerequisite to establish nuclear safeguards and for nuclear forensics techniques to prevent illicit trafficking of nuclear materials, methods to detect and characterize nuclear materials and trace their origins are required. The illicit nuclear activities of North Korea are ever increasing and raise warnings for the world. Hence, domestic concerns are being raised for the development of realistic detection technology and strengthening safeguards. To investigate the characteristics of nuclear materials, mass spectrometry is typically applied. Although mass spectrometry leads to very accurate analytical results, it has the drawbacks of high costs of the device and its operation involving a long process and destructive sample analysis [1,2]. Hence, a new method for obtaining analytical results in a shorter time and in a nondestructive manner is required; however, the research and development on this topic has been quite limited. According to the recommendations by the International Atomic Energy Agency (IAEA), one of the nuclear forensics requirements of the analytic procedure and time at the National Forensic Laboratory is that the U isotopic contents be identified within a day using  $\gamma$ -spectroscopy or  $\alpha$ -spectroscopy [3].

Domestically, a few studies have been performed to determine the U and Th contents [4] and the isotopic contents of U [5] by irradiating neutrons in a research reactor and to determine the U enrichment by gamma-ray spectroscopy using a commercial analytic software package [6]. In our preceding study on this topic [7], the so-called multigroup analysis code, HyperGam-U, was developed to analyze the  $XK_{\alpha}$  region in  $\gamma$ -X spectroscopic measurement, and a performance check was performed that involved identification of the  $^{235}$ U enrichment in eight uranium samples from certified reference materials (CRMs).

In the present study, a full analysis for the same U-spectra was performed to achieve a basic characterization of the sample and to develop the method and procedures. To assess the performance of the method, the contents and weight fractions of <sup>234</sup>U, <sup>235</sup>U, and <sup>238</sup>U in eight CRM samples with different <sup>235</sup>U enrichments in the range of 1–97% were determined and compared with the known values.

There are four methods used to date the time of purification and/or enrichment of U samples: i) determining the number ratio of  $^{230}$ Th/ $^{234}$ U by mass spectrometry [8–10], ii) determining the

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number ratio of  $^{231}$ Pa/ $^{235}$ U by high-resolution mass spectrometry [10], iii) determining the number ratio of  $^{232}$ Th/ $^{236}$ U for reprocessed U samples [11], or iv) determining the activity ratio of  $^{214}$ Bi/ $^{234}$ U by gamma spectroscopy [12,13].

Among these methods, the reported method iv), which is based on gamma spectroscopy, produces an accurate age; however, previous studies have used two types of hyper-pure germanium (HPGe) detectors, and the measurements are performed for 3 days in a low-background chamber [12,13]. The present study involves a new method of dating based on the activity ratio of <sup>223</sup>Ra/<sup>235</sup>U, which is extracted from a gamma spectrum acquired in 24 h and converted to the so-called "model age" based on the Bateman equation. The resulting ages are reviewed.

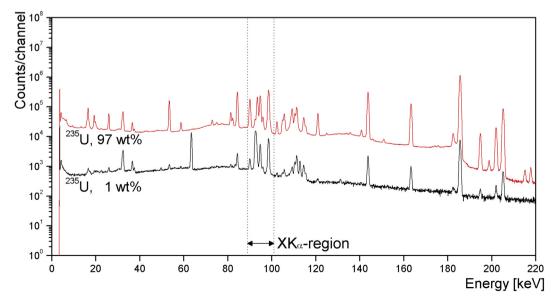
#### 2. Experiment

The samples used in the measurement were uranium CRMs with eight different enrichment levels in the range of 1–97%. These samples were manufactured in the New Brunswick Laboratory, and the contents are certified. These samples were used mainly for calibrating the mass spectrometers. Regarding the ages of these reference materials, only the ages of six samples were available. The containment and measurements of the samples were conducted in the Nuclear Chemistry Division of the Korean Atomic Energy Research Institute. In our preceding study, the  $UK_{\alpha}$  region of the spectrum was analyzed by a new code, HyperGam-U, and the resultant weight ratio of  $^{235}$ U/ $^{238}$ U was consistent within the relative error limit of 2% with the certified values [7]. The samples are highly pure U<sub>3</sub>O<sub>8</sub> powder. The exact masses are unknown, but they are less than approximately 1 g. Each sample was contained in a glass bottle of 1 mm thickness and 20 mm diameter. For the measurement, the sample was double caged in a polyethylene cylinder that is 1-mm thick for handling safety. The point reference sources for the energy and efficiency calibrations were placed in the same carriage and measured at the same distance from the detector endcap. The detector used was a medium-sized planar HPGe detector (GLP-36360; ORTEC, Oak Ridge, Tennessee, USA) in which the crystal's active volume was 13 mm (thickness) × 36 mm (diameter). and the endcap window was a Be window 0.254-mm thick. The detector has a resolution (full width at half maximum) of 585 eV for 122-keV gamma-rays. Both the detector and U sample were located in a lead cage 101-mm thick, and the distance was 5 cm between the sample's bottom plane and the endcap window. The measurements for each sample were conducted for the same live time of 24 h, and the dead time was below 3%.

#### 3. Analysis

Fig. 1 shows the gamma spectra of the CRM samples with enrichments 97% and 1% that were measured and analyzed in this study. The 89- to 101-keV region of the U spectrum is called the  $XK_{\alpha}$ -region, and there are approximately 13 mixed peaks from  $\gamma$ -decay and fluorescence X-rays emitted from <sup>235</sup>U, <sup>238</sup>U, and their daughter nuclei. In the preceding study of this work, the mixed peaks in the  $XK_{\alpha}$ -region were resolved by a multiplepeak fitting method, which is known as the multigroup analysis [14–17], and the number ratio of  $^{235}U/^{238}U$  was obtained. The result of the previous analysis was consistent with the certified values within the 2% relative error limit. However, the previous analysis did not assess the basic characterization of the samples. which includes the content of <sup>234</sup>U, U contents, and weight percents [7]. The 101- to 118-keV region of the spectrum is called the  $XK_{\beta}$ -region, which contains many  $\gamma$ - and X-ray peaks similar to the  $XK_{\alpha}$ -region. There exists no study that has resolved this XK<sub>β</sub>-region, and the region overlaps with the K-edge of the uranium attenuation factor at 115 keV, which complicates the analysis even further; thus, a suitable method must be developed. The information that can be obtained from the analysis of this region would be similar to that from the XK<sub>α</sub>-region; however, this is left for future investigations.

The peak search and analysis of the peak fitting and area determination have been performed by using the code HyperGam [18,19]. The analysis of the  $XK_{\alpha}$ -region was performed in the previous work [7], and the resultant peak areas of  $^{235}U$ ,  $^{238}U$ , and their daughters were obtained. The peaks in the  $XK_{\beta}$ -region were excluded from further analysis. The peaks detected in the other regions were identified for the emitting isotopes. The activities were calculated using the weighted mean of the peak activities, and the required nuclear data were from recent IAEA publication data [20]. The data for  $^{227}$ Th and  $^{230}$ Th were missing from this dataset and used from another dataset [21]. In the sample spectrum of an enrichment of 97%, more than 90 peaks were detected, whereas for



**Fig. 1.** The  $\gamma$ -ray energy spectra for the CRM uranium samples with 97% and 1% enrichment. CRM, certified reference material.

an enrichment of 1%, approximately 40 peaks were detected. In the spectra of the samples with mid-range enrichments, the number of the detected peaks was in between the two extremes. In Table 1, the gamma-ray peaks used for the present analysis are listed by the emitting nuclides and for the sample enrichment of 97%.

#### 3.1. Uranium contents

The absolute detection efficiency was determined by measuring the standard reference point sources. The calibrated efficiencies in the range of 60–160 keV were fitted by a second-order polynomial of  $logE_{\gamma}$ . All the samples in this study have a thickness of less than 300 mg/cm<sup>2</sup>, and self-attenuation was small. In the first analysis, therefore, the peak activities and their uncertainties were determined by using the efficiency based on the point reference sources. In the existing studies on uranium level structures or in the measurements for producing nuclear data, measurements on purified uranium or very fresh uranium were conducted to make the analysis of the measured spectrum comparatively simple. In the "infinite thickness method" [22,23], the uranium content is obtained by measuring the count rates of the 98.44-keV X-ray and the 185.7-keV gamma-ray. This method is applicable to and limited to samples of a given shape and density under the same detection conditions and hence severely limits the extraction of information associated with the many other peaks in the spectrum. Most of the samples in this study have aged 50 years or more since their purification and enrichment. As a result, the nuclides of <sup>235</sup>U, <sup>238</sup>U, <sup>234</sup>U, and their daughters are only partially radioequilibrated; the number of emitted gamma-rays is large, and most of the detected gamma peaks are doublets or multiplets. Therefore, a complete and comprehensive analysis of the spectrum peaks requires further time and effort. References on these aspects and useful analysis software are not available at the moment. Hence, the target of the present analysis is to set the data acquisition and analysis to 24 h. By using the peak activities, excluding those of the doublets or multiplets, all the data were averaged by the weights of inverse variance to obtain the nuclear activities. In the conversion, the latest IAEA decay data were used for the half-lives and emission probabilities [20], as shown in Table 2. The sample codes in Tables 2-6 contain three digit numbers which represent an approximate number of <sup>235</sup>U enrichment multiplied by 10. In addition to excluding the multiplets, the data for energies below 80 keV were also excluded because of the uncertainty in the efficiency of the point reference sources, and those for energies above 270 keV were also excluded because of the poor statistics from their low emission probabilities, interference from other peaks, and uncertain nuclear data. The specific activity of <sup>238</sup>U is low, and the gamma decay emission probabilities are also low: 49.55 keV (0.0697%) and 113.5 keV (0.0174%). The gamma peaks were detected in the 1% sample, but they were not in the 97% sample. Hence, the <sup>238</sup>U gamma decay peaks were not included in the analysis. Instead, the <sup>234</sup>Th activity was assessed by analyzing the 92.4- and 92.8-keV peaks in the  $XK_{\alpha}$ -region. This activity becomes approximately 97% of the <sup>238</sup>U activity 3 months after the sample

**Table 1** The  $\gamma$ -rays used to determine the activities of nuclides in the measured U-spectra.

Nuclide	Analyzed gamma-rays [keV]
<sup>235</sup> U	96.4, 140.7, 143.7, 150.9, 163.2, 182.4, 185.6, 194.8, 201.9,
	205.1, 215.0, 221.1, 228.6, 233.2, 240.6, 246.6, 266.1
<sup>231</sup> Th	84.3, 89.950, 89.957, 99.35, 124.9, 133.9, 135.6, 163.2, 174.0, 217.7
<sup>234</sup> Th	92.37, 92.79
<sup>234</sup> U	120.9
<sup>223</sup> Ra	269.13

**Table 2**Measured activities and contents of nuclides in the CRM samples (based on the efficiency of point reference sources).

-	Activity [Bq]			Mass [mg]		
code	<sup>235</sup> U	<sup>238</sup> U	<sup>234</sup> U	<sup>235</sup> U	<sup>238</sup> U	<sup>234</sup> U
U010	286 (7) <sup>a</sup>	4208 (165)	3840 (550)	3.6 (1)	338 (13)	0.017 (2)
U030	416 (9)	1907 (75)	5400 (620)	5.2(1)	153 (6)	0.023(3)
U050	1144 (22)		$1.29(14) \times 10^4$		256 (1)	0.056(6)
U100	1728 (33)	2164 (86)	$2.04(20)\times10^4$	21.6 (4)	174 (7)	0.088 (9)
U200	5243 (93)	3006 (120)	$6.31(62)\times10^4$	65.6 (12)	242 (10)	0.27(3)
U500	11750 (218)	1824 (73)	$2.97(40) \times 10^5$	147 (3)	147 (6)	1.3(2)
U850	29130 (500)	775 (36)	$4.54(51) \times 10^5$	364 (6)	62 (3)	2.0(2)
U970	45170 (770)	102 (8)	$1.19(12) \times 10^6$	565 (10)	8.2 (7)	5.2 (5)

CRM, certified reference material.

**Table 3**Comparison of the weight contents (based on the efficiency of point reference sources) with those of certified reference values.

-	This analysis <sup>a</sup> [w%]			Certified reference values <sup>b</sup> [w%]		
code	<sup>235</sup> U	<sup>238</sup> U	<sup>234</sup> U	<sup>235</sup> U	<sup>238</sup> U	<sup>234</sup> U
U010	1.05 (5) <sup>c</sup>	98.95 (547)	0.005 (1)	0.9911	98.997	0.00532
U030	3.28 (14)	96.70 (531)	0.015(2)	3.009	96.953	0.0187
U050	5.30 (23)	94.68 (514)	0.021(2)	4.949	94.975	0.0275
U100	11.05 (45)	88.91 (472)	0.045 (5)	10.075	89.821	0.0666
U200	21.33 (77)	78.59 (399)	0.089(9)	19.811	79.856	0.1229
U500	49.84 (144)	49.73 (228)	0.438 (60)	49.383	50.029	0.5126
U850	85.01 (200)	14.53 (71)	0.460 (52)	84.988	14.001	0.6399
U970	97.69 (234)	1.42 (12)	0.893 (89)	97.663	0.5296	1.6582

<sup>&</sup>lt;sup>a 236</sup>U could not be analyzed in this study.

**Table 4** Measured activities and contents of nuclides in the CRM samples (based on the efficiency for  $U_3O_8$  massive sources).

-	Activity [Bq]	Mass [mg]				
code	<sup>235</sup> U	<sup>238</sup> U	<sup>234</sup> U	<sup>235</sup> U	<sup>238</sup> U	<sup>234</sup> U
U010	289 (7) <sup>a</sup>	4267 (168)	3880 (560)	3.6(1)	343 (13)	0.017 (2)
U030	416 (9)	1909 (75)	5400 (620)	5.2(1)	154 (6)	0.023(3)
U050	1151 (23)	3208 (126)	$1.30(14)\times10^4$	14.4(3)	258 (1)	0.056(6)
U100	1731 (33)	2172 (86)	$2.04(20)\times10^4$	21.6 (4)	175 (7)	0.089(9)
U200	5287 (95)	3041 (121)	$6.37(63)\times10^4$	66.1 (12)	245 (10)	0.28(3)
U500	11840 (220)		$2.99(41)\times10^{5}$		148 (6)	1.3(2)
U850	29590 (510)	790 (36)	$4.62(52) \times 10^5$	370 (6)	64(3)	2.0(2)
U970	46250 (790)	105 (9)	$1.22 (12) \times 10^6$	579 (10)	8.5 (7)	5.3 (5)

CRM, certified reference material.

Comparison of the weight contents (based on the efficiency for  $U_3O_8$  massive sources) with those of certified reference values.

codo	This analysis [w%]			Certified reference values [w%]		
	<sup>235</sup> U	<sup>238</sup> U	<sup>234</sup> U	<sup>235</sup> U	<sup>238</sup> U	<sup>234</sup> U
U010	1.04 (5) <sup>a</sup>	98.95 (547)	0.005 (1)	0.9911	98.997	0.00532
U030	3.28 (14)	96.71 (531)	0.015(2)	3.009	96.953	0.0187
U050	5.29 (22)	94.69 (514)	0.021(2)	4.949	94.975	0.0275
U100	11.03 (44)	88.93 (472)	0.045 (5)	10.075	89.821	0.0666
U200	21.27 (77)	78.64 (399)	0.089 (9)	19.811	79.856	0.1229
U500	49.76 (144)	49.81 (229)	0.438 (60)	49.383	50.029	0.5126
U850	84.95 (200)	14.59 (71)	0.461 (52)	84.988	14.001	0.6399
U970	97.67 (234)	1.43 (12)	0.896 (89)	97.663	0.5296	1.6582

 $<sup>^{\</sup>text{a}}\,$  The number in the parenthesis is  $1\sigma$  uncertainty of the preceding number.

<sup>&</sup>lt;sup>a</sup> The number in the parenthesis is  $1\sigma$  uncertainty of the preceding number.

 $<sup>^{\</sup>rm b}$  Values for  $^{236}$ U are obtained in the certified reference sheets but are not listed in this table.

 $<sup>^{\</sup>text{c}}\,$  The number in the parenthesis is  $1\sigma$  uncertainty of the preceding number.

 $<sup>^{\</sup>rm a}$  The number in the parenthesis is  $1\sigma$  uncertainty of the preceding number.

 Table 6

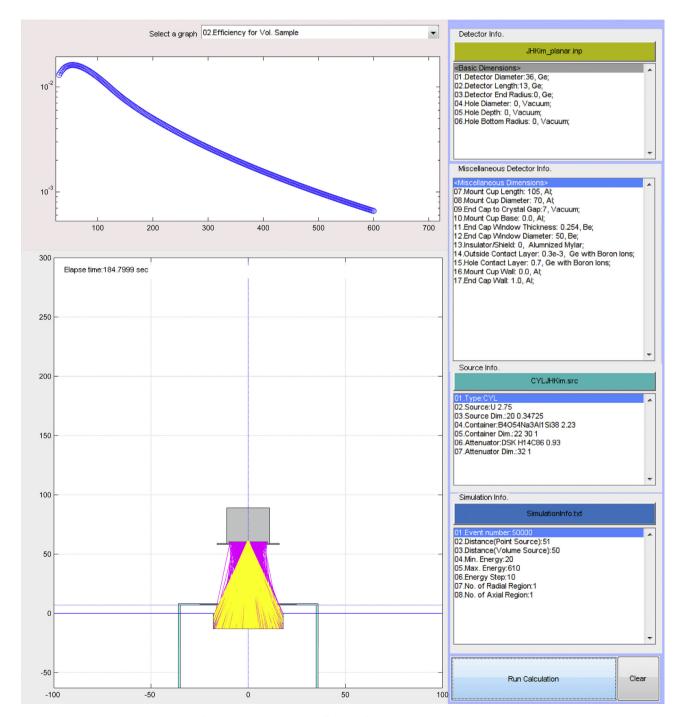
 The measured model ages in this study compared with the true ages of the CRM.

Sample code	U970	U850	U500	U200
True age (yr)	49.3	56.5	55.5	N.A.
Model age (yr)	62 ± 4	62 ± 4	58 ± 5	51 ± 8

CRM, certified reference material.

purification. Because the ages of all the present samples are more than several decades, the <sup>238</sup>U activity was considered to be the same as that of <sup>234</sup>Th. The activity of <sup>234</sup>U was obtained from the 120.9-keV (0.0386%) peak. In Table 3, the composition (weight

percent, w%) obtained in this work is compared with those of the certified reference values. For the  $^{235}$ U content of the U970 sample, the relative bias (%), which is given as [this analysis—certified value]/[certified value]  $\times$  100, is accurate to +0.03%, and for the U010 sample, it is accurate to +5.9%. Although the contents of  $^{238}$ U exhibit large statistical error, they are consistent with the certified values. The content of  $^{234}$ U determined is consistent within the error range for the low enrichment samples, whereas those determined for the high enrichment samples outranged the uncertainties. These issues require an investigation of additional causes, and the priority work would be a longer measurement to improve the inherently low statistics.



**Fig. 2.** The EXVol window shown for a simulation of  $\gamma$ -rays to calculate the peak efficiencies for the CRM uranium samples. CRM, certified reference material.

In the second stage of the analysis, the absolute detection efficiencies were calculated by using the U contents obtained in the first step and by taking the U sample as a volume source with a thin disc shape. The peak efficiency for a volume source is, according to Moens' concept of an effective solid angle [24], given by

$$\varepsilon_{vol} = (\Omega_{geo}/4\pi) F_{att} \varepsilon_p^{int} \tag{1}$$

where  $\Omega_{geo}$  is the geometric solid angle,  $F_{att}$  is the gamma-ray's attenuation factor for the media in the path between the source position and the entrance position of the detector crystal, and  $\varepsilon_p^{int}$  is the intrinsic peak efficiency of the detector. The corresponding quantities are given for the reference point source in Eq. (1). Here, the principle is applied so that the intrinsic peak-to-total ratio is independent from the position of the source:  $\frac{\varepsilon_{p,rol}^{int}}{\varepsilon_{r,ol}^{int}} = \frac{\varepsilon_{p,rol}^{int}}{\varepsilon_{r,ol}^{int}}$  [24]. The intrinsic peak efficiency for a volume source is therefore given by the measured peak efficiency for the reference point source and the ratio of the calculated quantities of the geometric solid angle, attenuation factor, and intrinsic total efficiency for both sources [24,25]:

$$\varepsilon_{vol} = \varepsilon_{ref} \left( \Omega_{geo} F_{att} \varepsilon_t^{int} \right)_{vol} / \left( \Omega_{geo} F_{att} \varepsilon_t^{int} \right)_{ref}. \tag{2}$$

To calculate the quantities, the code EXVol was used. In the EXVol calculation, each factor in Eq. (2) is obtained by simulating the gamma-ray trajectories using the Monte Carlo method and calculating the attenuation factors and path lengths along the path. Fig. 2 shows the simulated effective solid angle of the point reference source for the detection geometry and the EXVol window for calculating the efficiency for the U source in this study. The detection efficiency for the U sources in this study is given in Fig. 3 according to the  $\rm U_3O_8$  masses. The EXVol code can exactly treat the K-absorption edge of U attenuation factor at 115 keV [26]. Because the mass thicknesses of the U samples in this study are small, the K-edge is not apparent from the calculated efficiencies. As shown in

Fig. 3, the effects caused by the sample's solid angle, self-attenuation, and detection efficiency are within 10% for the U source masses of less than 1 g in this study, and a new analysis was performed for the U isotopic activity and content by using the mass of each sample. In Table 4, the renewed isotopic activities and contents are shown. In Table 5, the weight contents (weight percent, w%) are compared with those of the certified reference values. The analytical results in Tables 4 and 5 have been improved within ca. 5% for the nuclide activities and mass contents in comparison with the corresponding quantities in Tables 2 and 3, which are based on the point source efficiency.

#### 3.2. Age dating

The method used in this study for age dating is based on the ratio of activities of  $^{235}$ U and its daughter  $^{223}$ Ra. The sample's age is the time that has passed since the last reprocessing, chemical separation, or enrichment to the moment of the measurement. To convert the number ratio or activity ratio into the age, the Bateman equation [27] is used; therefore, this age is called the "model age." This age is the number based on the assumption that the chemical separation of uranium is perfect at time t = 0, and hence, the number of daughter nuclei is zero. This condition leads to an equation for the daughter product activity at time t in terms of the  $^{235}$ U's activity at time t=0 or at time t. When the number ratio or the activity ratio is determined by the experimental method and the equation for the age t, given the ratio, is solved, the age is obtained. Conventionally, the number ratio of <sup>235</sup>U/<sup>231</sup>Pa is determined by mass spectrometry [10], and its signal is higher by a maximum of  $10^3$  than that based on the ratio of  $^{235}\text{U}/^{223}\text{Ra}$ . In this study, we searched the possible candidate gamma lines when the activity ratio of <sup>235</sup>U/<sup>231</sup>Pa is determined by gamma spectrometry. Among the gamma lines of <sup>231</sup>Pa having an emission probability greater than 1%, there are four gamma lines (27.37, 283.69, 300.06, and 302.67 keV) [20] that are detectable by the spectrometer of this study. The 27.37-keV gamma line has the highest emission probability and the largest peak. The peak is, however, a doublet mixed

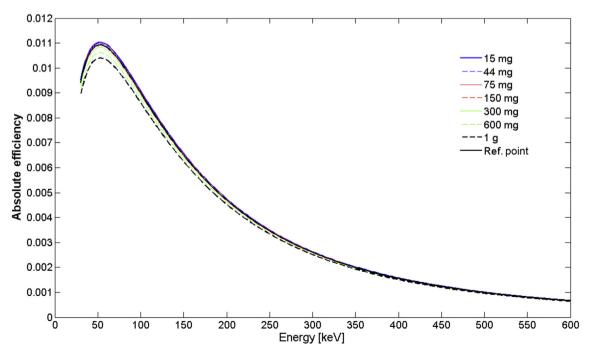
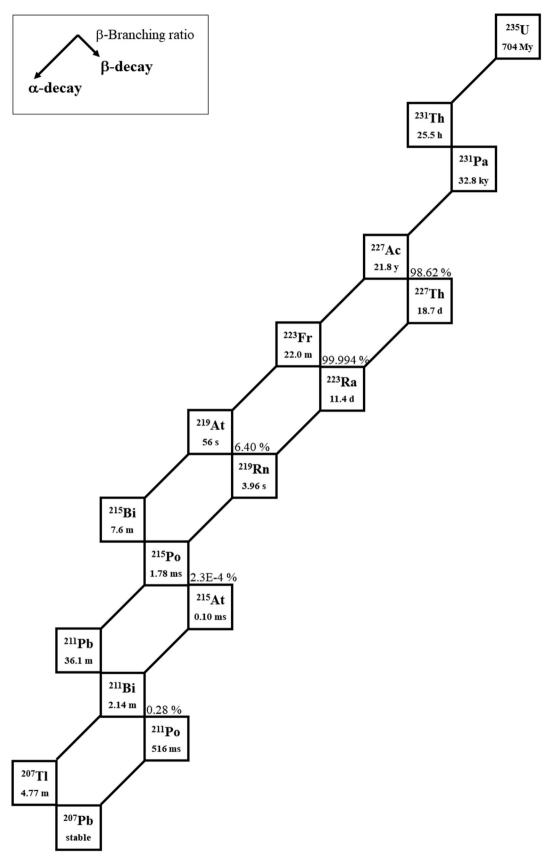


Fig. 3. The absolute peak efficiencies for the U<sub>3</sub>O<sub>8</sub> powder samples of various masses in this study.



**Fig. 4.** The decay chain of  $^{235}$ U.

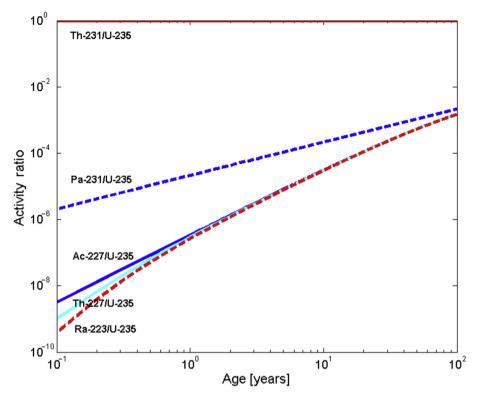


Fig. 5. The ratio of activities between <sup>235</sup>U and its daughter products.

with a gamma peak of <sup>231</sup>Th; this mixing makes resolving these peaks impossible. All the other peaks are not detected within 24 h of counting. Then, the method based on the  $^{235}\text{U}/^{231}\text{Pa}$  ratio is inappropriate for this study. Therefore, the activity ratios of <sup>235</sup>U/ <sup>223</sup>Ra were obtained in the spectra analyses, and hence, the model ages of the samples were determined in this study. The peak to determine the activity of <sup>223</sup>Ra is the 269.5-keV gamma peak, and its emission probability is 14.23 (32)%, i.e., the highest emitting peak of <sup>223</sup>Ra [20]. All the other lines of <sup>223</sup>Ra are mixed with the gamma peaks of other nuclides, making the analysis complicated or impossible. The activity of <sup>235</sup>U was obtained in the previous procedure, as shown in Table 2. In Fig. 4, the decay chain of <sup>235</sup>U is shown. When the age exceeds several months, the activity of <sup>223</sup>Ra approaches that of  $^{227}$ Ac, as shown in Fig. 4. The  $^{231}$ Pa activity becomes asymptotically  $^{22}$  ×  $^{235}$ U's activity after 100 years. At age t, the activity of  $^{223}$ Ra is given by the following expression, where the subscript of each term denotes the corresponding term: 1,  $^{235}$ U; 2,  $^{231}$ Th; 3,  $^{231}$ Pa; 4,  $^{227}$ Ac; 5,  $^{227}$ Th; and 6,  $^{223}$ Ra. Here,  $\lambda_i$  is the decay constant of the nuclide, and N<sub>i</sub>(t) is the number of the nuclide at time t:

$$N_{6}(t) = N_{1}(0)\lambda_{1}\lambda_{2}\lambda_{3}\lambda_{4}\lambda_{5}\sum_{j=1}^{6}D_{6j}\exp\left(-\lambda_{j}t\right) \tag{3}$$

where

$$D_{6j} = \begin{cases} 1, & i = j \\ 1 / \prod_{i=1}^{6} (\lambda_i - \lambda_j), & i \neq j \end{cases}$$
 (4)

The activity ratios of a daughter product and that of the parent <sup>235</sup>U were calculated and are shown in Fig. 5 according to the age t. Eqs. (3) and (4) are given in terms of age t. To obtain the age t, a numerical iterative process was adopted for the given ratio of the

<sup>223</sup>Ra/<sup>235</sup>U activities. The measurement time in this study was July 2014. For this date, the obtained model ages and the known true ages were compared in Table 6. In Table 6. N.A. denotes that the sample's true age was not available in terms of traceable documents. For the four samples of enrichments less than 40%, the <sup>223</sup>Ra 269.5-keV peak was not detected in the present measurements. This lack of detection could be caused by the low <sup>235</sup>U activity of the 40% sample, which requires a longer counting time and/or a larger quantity of the sample. The model age of the 50% enriched sample reproduced the true age, whereas the model ages of the other samples overestimated the ages as the enrichment increased. The reasons for this inconsistency and trend are either i) the statistics of the present detection are not sufficient because it is a 24-h data acquisition or ii) the chemical separation of uranium was incomplete at the initial point in some of the samples. Hence, the sample of 97% enrichment was detected for a longer period of 10 days, and the ratio of <sup>223</sup>Ra/<sup>235</sup>U activities was converted to a model age of  $56 \pm 2$  year, which was closer to the true age of 49.3 year. However, the result remained beyond the uncertainty value. Another influence on the bias could be traced to a study that report that enriched uranium obtained in the early years of developing enrichment techniques may contain impurities caused by premature chemical separations [9]. It is not appropriate to identify the reason based on the present study of a few samples. A further study is required in the future.

# 4. Conclusion

A method and codes were developed to determine the total U content, isotopic content, and the enrichment of <sup>235</sup>U based on high-resolution gamma spectrometry, and then, its performance was investigated by applying it to U CRMs. The eight different CRM samples of enrichments in the range of 1–97% were counted for 24 h, and the obtained gamma spectra were analyzed. The peaks in

the  $XK_{\alpha}$  region were analyzed by using the new code HyperGam-U, and the peaks in the other regions were analyzed using the code HyperGam. The code EXVol was used to obtain the absolute peak efficiency for the uranium sources based on the volume. In the peak analysis, the activity ratio of  $^{235}\text{U}/^{223}\text{Ra}$  was determined, and a code based on solving the Bateman equation was developed; the model ages of the enriched samples were obtained and compared with the true ages. The present study showed that a basic characterization in terms of the total U content and the contents of  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$  in a uranium sample is feasible using a 24-hour detection period on gram or subgram samples. In addition to the main characterization, information on the sample's model age was acquired by using the developed method and code.

#### **Conflicts of interest**

All authors have no conflicts of interest to declare.

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