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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_{α} 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.

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 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 γ-spectroscopy or α-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_{α} region in γ-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 ^{234}U, ^{235}U, and ^{238}U in eight CRM samples with different ^{235}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 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 $^{223}Ra/^{235}U$, 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_α 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_3O_8 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_α-region, and there are approximately 13 mixed peaks from γ-decay and fluorescence X-rays emitted from ^{235}U, ^{238}U, and their daughter nuclei. In the preceding study of this work, the mixed peaks in the XK_α-region were resolved by a multiple-peak 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 ^{234}U, U contents, and weight percents [7]. The 101- to 118-keV region of the spectrum is called the XK_β-region, which contains many γ- and X-ray peaks similar to the XK_α-region. There exists no study that has resolved this XK_β-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_α-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_α-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_β-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

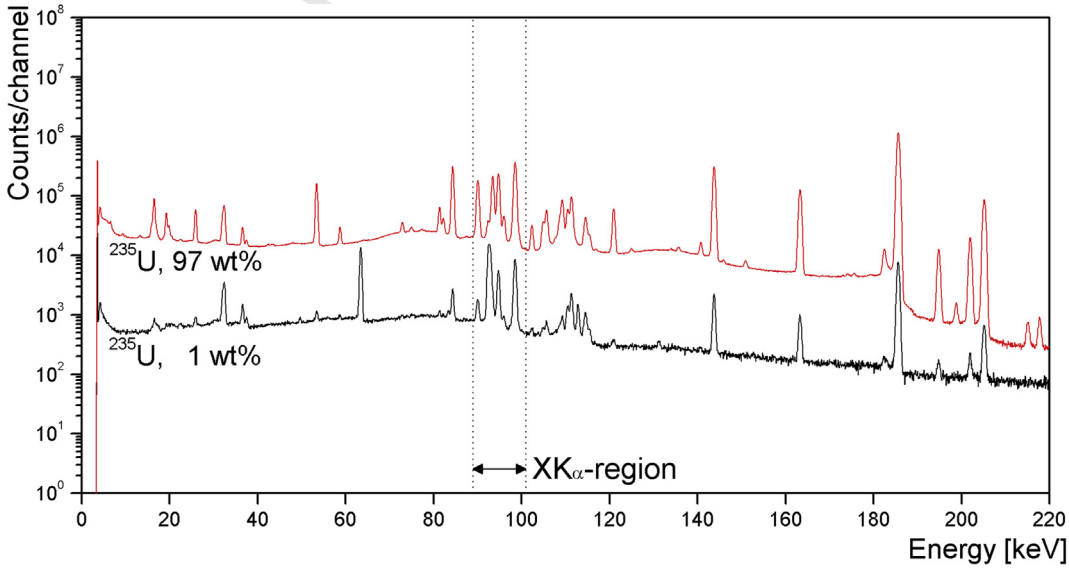

Fig. 1. The γ-ray energy spectra for the CRM uranium samples with 97% and 1% enrichment.
CRM, certified reference material.

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