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Original Article

Uranium Enrichment Determination Using a New Analysis Code for the U XK_α Region: HyperGam-U

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ABSTRACT

HyperGam-U was recently developed to determine uranium enrichment based on γ- and X-ray spectroscopy analysis. The XK_α region of the uranium spectrum contains 13 peaks for ^{235}U and ^{238}U and is used mainly for analysis. To describe the X-ray peaks, a Lorentzian broadened shape function was used, and methods were developed to reduce the number of fitting parameters for decomposing the strongly overlapping peaks using channel-energy, energy-width, and energy-efficiency calibration functions. For validation, eight certified reference material uranium samples covering uranium enrichments from 1% to 99% were measured using a high-resolution planar high-purity germanium detector and analyzed using the HyperGam-U code. When corrections for the attenuation and true coincidence summing were performed for the detection geometry in this experiment, the goodness of fit was improved by a few percent. The enrichment bias in this study did not exceed 2% compared with the certified values for all measured samples.

1. Introduction

Uranium enrichment is one of the most important characteristics of nuclear materials, and needs to be assessed rapidly and accurately to strengthen nuclear nonproliferation and safeguards. Although mass spectroscopy is the most accurate method for analyzing uranium enrichment, a rapid nondestructive analysis method is required in the field. X- and γ-rays can be measured to determine the uranium enrichment of a sample nondestructively and can be performed easily without the time delay caused by transporting samples to the laboratory.

The measurement of 185.7 keV γ-rays emitted from ^{235}U, the so-called "infinite-thickness method," is a simple enrichment determination method. However, it requires enrichment-count rate calibration under accurate control of the detection geometry and using certified uranium standards with different enrichments and collimators. This method is suitable for routine analysis, such as measuring nuclear fuel casks in nuclear fuel factories or storage. However, it is

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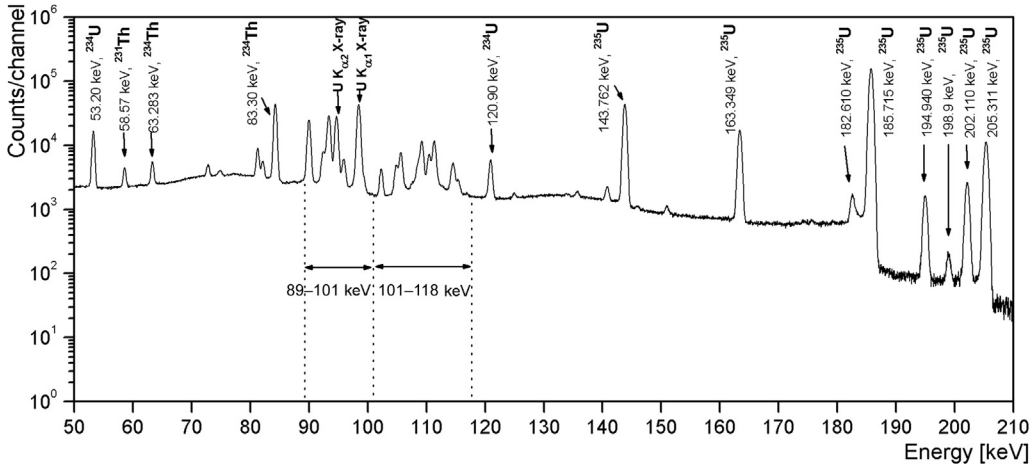

Fig. 1 – The 50–210 keV energy region of the uranium spectrum for a 50% enriched CRM uranium sample. CRM, certified reference material.

practically impossible to prepare reference samples with diverse chemical compositions and geometries for nuclear forensics or inspection at ports.

Gunnink [1], Sampson and Kelley [2], and Morel et al [3] developed a method that uses the XK_α region of the uranium spectrum, the so-called "intrinsic efficiency method." The XK_α region is useful for isotopic analysis because there is little variation in parameters such as the detection efficiency and peak resolution with photon energy in this narrow energy region. Studies have used the program HyperGam to analyze the high-purity germanium (HPGe) γ-spectra [4,5]. To analyze the uranium XK_α region, however, a new algorithm is required to describe the X-ray peaks and to reduce the number of fitting parameters. To measure the strongly overlapping peaks in the XK_α region, a planar HPGe detector was introduced for high-resolution spectroscopy.

Differences in the detection geometry, i.e., the chemical composition, density, and size of uranium samples, and the material and thickness of the sample container, alter the attenuation of γ- and X-rays. In addition, the true coincidence summing (TCS) effect results in a counting loss for the peaks for γ-ray cascades with a close counting geometry. Since these corrections for attenuation and the TCS effect are quite complicated for a volume source geometry, this study performed simpler corrections for the measurements.

2. Materials and methods

2.1. *Uranium XK_α region*

The 50–210 keV energy region of the uranium spectrum obtained with a planar HPGe detector is shown in Fig. 1. The 89–101 keV energy region, the so-called XK_α region, contains 13 useful γ- and X-ray peaks for ^{235}U and ^{238}U. Enrichment of uranium samples can be determined from an analysis of this region. The 101–118 keV region has a similar overlapping structure, but it is less useful because of the low photon emission probabilities of ^{238}U and its daughters (less than 0.04% each). Moreover, little is known of the emission probabilities of thorium and protactinium X-rays tied to ^{235}U decay.

When secular equilibrium of a uranium sample is achieved, the activity of the uranium equals that of its daughter nuclides. Hence γ- and X-rays can be classified into three component groups by their origin and progeny: ^{235}U and its daughters, ^{238}U and its daughters, and fluorescence X-rays.

2.2. *Spectrum fitting*

The fitting process is performed with empirical formulae representing the peak main, exponential tails, and background functions. Fitted values at channel J are given as follows:

$$F(J) = \sum_{j,k}\left\{f_{j,k}(J) + ST_{j,k}(J) + LT_{j,k}(J) + STB_{j,k}(J)\right\} + SB(J) \quad (1)$$

where $f_{j,k}$ is the main part function of the j^{th} peak of the k^{th} component ($k = 235$, 238, and F for U^{235}, U^{238}, and fluorescence X-rays, respectively), $ST_{j,k}$ is the short-term tail function of the j^{th} peak, $LT_{j,k}$ is the long-term tail function of the j^{th} peak, $STB_{j,k}$ is the step background function of the j^{th} peak, and SB is the function of the smooth background formed along the entire analysis energy region. The shape of the γ- or X-rays is mainly described by the Gaussian or Lorentzian broadened shape, respectively:

$$f_G(x) = H\cdot\exp\left(-\frac{x^2}{\delta^2}\right) \quad (2)$$

$$f_X(x) = H\cdot K\left(\frac{x}{\delta}, \frac{\Gamma}{2\delta}\right) \quad (3)$$

$$K(x,y) = \frac{y}{\pi}\int_{-\infty}^{+\infty}\frac{\exp(-\theta^2)}{(x-\theta)^2 + y^2}d\theta \quad (4)$$

where x is the channel location compared to the peak center $P_{j,k}$ ($x = J - P_{j,k}$), H is the peak height, δ is the peak width