Determination of 235U/238U atom ratio by low energy photon spectrometry with a silicon drift detector

Authors: Dr. Haifei Zhang, Chen, Dr. Wei, Xiao-Long Huang, Yang, Prof. Dong, Dr. Haifei Zhang

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Abstract

A methodology based on low energy photon spectrometry with a silicon drift detector (SDD) is developed for the determination of 235U/238U atom ratio in uranium samples. The SDD is well suited for measuring low energy photons emitted from thin uranium sample source with low self-absorption. The main advantage of this method is that it allows for a direct determination of the U isotope ratio, while requiring little sample preparation and less labor intensive than methods requiring radiochemical treatment. The obtained mixed X-ray and -ray spectrometry for isotopic analysis of 235U/238U are investigated from 13.0, 13.3, 13.56, 15.6, 16.2, 16.78, 17.17 and 19.58 keV photons.

Full Text

Preamble

A methodology based on low-energy photon spectrometry with a silicon drift detector (SDD) is developed for determining the 235U/238U atom ratio in uranium samples. The SDD is well suited for measuring low-energy photons emitted from thin uranium sample sources with minimal self-absorption. The main advantage of this method is that it enables direct determination of the uranium isotope ratio while requiring little sample preparation and being significantly less labor-intensive than methods requiring radiochemical treatment. The mixed X-ray and -ray spectrometry for isotopic analysis of 235U/238U is investigated using photons at 13.0, 13.3, 13.56, 15.6, 16.2, 16.78, 17.17, and 19.58 keV.

Keywords: 235U/238U atom ratio, low-energy photon spectrometry, silicon drift detector

Introduction

Natural uranium consists of a mixture of three isotopes: 238U (99.2742%), 235U (0.7204%), and 234U (0.0054%) [1, 2]. Uranium is enriched in the 235U isotope to enhance its fissile characteristics for use as nuclear fuel. Based on the 235U/238U atom ratio, enriched uranium can be classified as high-enriched uranium (more than 20% 235U) and low-enriched uranium (less than 20% 235U), with a variant of slightly-enriched uranium (between 0.9% and 2% 235U) [2, 3]. Determination of the 235U/238U atom ratio is required for various fields, including radiation protection and nuclear energetics.

The 235U/238U atom ratio r can be defined by the simple formula:

$$r = \frac{N(^{235}\mathrm{U})}{N(^{238}\mathrm{U})} = \frac{A(^{235}\mathrm{U})}{A(^{238}\mathrm{U})} \cdot \frac{T_{1/2}(^{235}\mathrm{U})}{T_{1/2}(^{238}\mathrm{U})}$$

where A is the activity and $T1/2$ is the half-life of the respective nuclide.

The determination of the 235U/238U atom ratio can be implemented by nuclear methods such as alpha or gamma spectrometry [4, 5] or by mass spectroscopic techniques such as Inductively Coupled Plasma Mass Spectrometry (ICP-MS) [6, 7]. Mass spectrometry has long been the benchmark technique for this purpose since it provides the most precise results. However, this method involves laborious sample preparation and is expensive. On the other hand, high-resolution gamma spectrometry using high-purity germanium (HPGe) detectors is fast and cost-effective, while its maintenance and in-situ deployment are laborious. Therefore, we investigated the determination of the 235U/238U atom ratio using a portable Peltier-cooled silicon drift detector (SDD) by employing low-energy photon spectrometry (X- and -ray peaks in the XK/L region). The main advantage of this method is that it allows for direct determination of the uranium isotope ratio while requiring minimal sample preparation and being significantly less labor-intensive than methods requiring radiochemical treatment and maintenance. Furthermore, analysis of X- and -ray peaks in the XK/L region has the advantage of not requiring calibration with known enrichment standards, because the energy range of this region is limited and consequently the variation in detector efficiency response is small [8].

238U undergoes alpha decay ($T1/2 = 4.7 \times 10\hat{}9$ y) to daughter thorium-234, which immediately decays ($T1/2 = 24.1$ d) to protactinium-234m by beta-particle emission. The half-life of 234mPa is short (1.17 min) for transition to ground-state 234Pa. Then 234Pa decays ($T1/2 = 6.67$ h) to 234U by beta-particle emission. Therefore, the radionuclides 234Th, 234mPa, and 234Pa should rapidly reach secular equilibrium with parent nuclei [9]. 235U undergoes alpha decay ($T1/2 = 7.04 \times 10\hat{}8$ y) to daughter thorium-231. 231Th, due to its short half-life ($T1/2 = 25.5$ h), also establishes secular equilibrium with parent 235U within a reasonable time period. Since 235U and 238U do not emit adequately significant photons that can be used for low-energy ray spectroscopic determination, they are usually determined through their daughter products in equilibrium, namely 234Th, 234mPa, 234Pa, 234U, and 231Th. Therefore, the 235U/238U atom ratio is evaluated based on the nuclear decay data of 234Th, 234mPa, 234Pa, 234U, and 231Th.

Experimental Procedure

A low-energy photon spectrometer—Peltier-cooled silicon drift detector (SDD) from AMPTEK with a resolution of 125 eV (FWHM) at the 5.9 keV peak of 55Fe—was used for low-energy photon spectrometric analysis. It has a sensitive area of 25 mm^2, depleted thickness of 500 µm, and beryllium window of 0.5 mil. The detection apparatus is illustrated in [Figure 1: see original paper].

The main low-energy peaks can be identified from 235U, 238U, and daughter nuclides 231Th, 234Th, 234mPa, 234Pa, as shown in [Figure 2: see original paper]. The spectrum of 241Am was used for energy calibration. The peak at 13.0 keV corresponds to /X-ray transitions of 231Th and 238U, the peak at 13.3 keV corresponds to /X-ray transitions of 231Th and 234Th, the peak at 13.56 keV corresponds to the X-ray transition of 234Pa, the peaks at 15.6 keV and 16.2 keV correspond to the L-shell and M+-shell X-ray transitions of 231Th, the peaks at 16.78 keV and 17.17 keV correspond to the X-ray transitions of 238U, and the peak at 19.58 keV corresponds to /X-ray transitions of 235U and 231Th. The absolute intensities are calculated based on ENSDF [10] and listed in .

As associated electronics, the detector has an internal preamplifier and is attached, through multiple connections, to a custom module for detector control, voltage power supply, pulse amplifying and shaping, and ADC conversion. Digital data are fed to a multichannel analyzer and spectrum analysis ORTEC GAMAVISION-32 software via an Ethernet connection to a PC.

Results and Discussion

Initially, a small surface source of 241Am was used for energy calibration. After implementing an appropriate shaping process, the measured spectrum of 241Am is shown in [Figure 2: see original paper]. Following analysis, the peaks located at Channel 321 and Channel 714 correspond to 13.9 keV and 17.8 keV, with energy resolutions (FWHM) of about 0.20 keV and 0.23 keV, respectively.

Consequently, a piece of uranium source sample was measured. In the low-energy region of the uranium spectrum, several -ray and X-ray peaks are superimposed ([Figure 3: see original paper]). This figure reports the main energies of the peaks: those at 13.0, 13.3, 13.56, 15.6, 16.2, 16.78, 17.17, and 19.58 keV are complex ones.

Since the half-lives of 235U and 238U are much longer than those of all their daughter nuclides, two secular equilibrium chains exist: the 235U chain, denoted by Ch1, and the 238U chain, denoted by Ch2. At secular equilibrium, the activity of each nuclide in a chain is the same [11]. Therefore, the activity of 235U and each of its daughters can be considered equal, and we can set the activity relation as $A(235\text{U}) = A(231\text{Th}) = x$. Similarly, we can set the activity of 238U as $A(238\text{U}) = A(234\text{Th}) = A(234\text{Pa}) = y$. The uranium isotope activities can be resolved as follows:

where E /X is the energy value of a /X-ray in keV, I /X,i is the absolute intensity of the /X-ray emitted from the i-th nuclide in equilibrium chain Ch1, and I /X,j is the absolute intensity of the /X-ray emitted from the j-th nuclide in equilibrium chain Ch2. (E /X) is the detection efficiency of a /X-ray with energy E /X.

From AMPTEK FastSDD data, the intrinsic full-energy efficiency for our detection apparatus can be fitted by the following expression:

$$\varepsilon(E) = 0.0003E^3 - 0.0137E^2 + 0.1251E + 0.774$$

where E denotes the energy value of a /X-ray in keV, and (E) is the intrinsic efficiency. Based on Eq. (3), the intrinsic efficiencies for the eight peaks can be evaluated and are listed in . In this work, the intrinsic efficiency dominates the detection efficiency because the energy range of this region is limited and consequently the variation in detector efficiency response is small. This can be explained by the fact that, for processes measuring uranium enrichment based on analysis of the low-energy ray spectral region within the same geometrical factor Ω (Ω being the solid angle), the photon emission probabilities characterizing the disintegration of 235U and 238U are used in a relative manner.

For spectral deconvolution, the peak shapes were assumed to follow a Gaussian distribution with appropriate parameters. The continuum spectrum underneath the peaks was subtracted using a linear background. Using the ORTEC GAMAVISION-32 software for spectrum analysis, the net area and its corresponding statistical fluctuation for each peak can be obtained and are given in .

For the 13.6 keV peak, there is no X-ray transition in the 235U chain, so this peak is omitted from Eq. (2). In addition, there is limited or no nuclear data information for the 16.78 keV and 17.17 keV peaks, and their inclusion would introduce unpredictable effects; therefore, these two peaks are also omitted from Eq. (2). The nuclear databases for the 13.6 keV, 16.78 keV, and 17.17 keV /X-rays should be complemented in the future, as this represents decisive fundamental research for low-energy photon spectrometry. This leaves five equations in Eq. (2).

Finally, we substitute the net area values and intrinsic full-energy efficiencies into Eq. (2), obtaining the following system of equations:

$$\begin{aligned} 0.19 \cdot 0.743 \cdot x + 0.0247 \cdot 0.743 \cdot y &= 17498, \\ 0.24 \cdot 0.719 \cdot x + 0.0263 \cdot 0.719 \cdot y &= 18164, \\ 0.0473 \cdot 0.529 \cdot x + 0.0107 \cdot 0.529 \cdot y &= 5221, \\ 0.159 \cdot 0.479 \cdot x + 0.0786 \cdot 0.479 \cdot y &= 33259, \\ 0.056 \cdot 0.221 \cdot x + 0.0096 \cdot 0.221 \cdot y &= 1970. \end{aligned}$$

Using the least squares method to solve Eq. (4), we obtain $x = 11393$ and $y = 865703$. The 235U/238U atom ratio r can then be calculated:

$$r = \frac{A(^{235}\mathrm{U})}{A(^{238}\mathrm{U})} \cdot \frac{T_{1/2}(^{235}\mathrm{U})}{T_{1/2}(^{238}\mathrm{U})} = \frac{11393}{865703} \cdot \frac{7.04 \times 10^8}{4.7 \times 10^9} = 0.198\%.$$

The uncertainty r for the 235U/238U atom ratio r is given by:

$$\sigma_r = \sqrt{\sigma_c^2 + \sigma_f^2}$$

where c is the counting and peak analysis uncertainty estimated by the spectrum analysis software (including counting statistical uncertainty as well as uncertainty from peak fitting and deconvolution where applicable), and f is the uncertainty due to calibration curve fitting estimated during the fitting process. The uncertainty is then substituted into the uncertainty propagation formula to give the combined standard uncertainty. Counting uncertainty is less than 7.1%, peak analysis uncertainty is less than 5.5%, and calibration curve fitting uncertainty for Eq. (3) is less than 5.8%. The combined standard uncertainty r is therefore less than 10.7%.

Summary

The results of this work demonstrate that low-energy photon spectrometry with a silicon drift detector (SDD) can be applied for determining the 235U/238U atom ratio in uranium samples. Using X-ray and -ray peaks at 13.0, 13.3, 13.56, 15.6, 16.2, 16.78, 17.17, and 19.58 keV for analysis of the 235U/238U atom ratio is reasonable and applicable. Furthermore, it should be noted that very little sample preparation is required and the analysis is non-destructive. The nuclear databases for the 13.6 keV, 16.78 keV, and 17.17 keV /X-rays of the 235U/238U chains should be complemented in future work.

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Note: Figure translations are in progress. See original paper for figures.