Nuclear Instruments and Methods in Physics Research B 359 (2015) 137–144

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Gamma-spectrometric determination of ^{232}U in uranium-bearing materials

Jozsef Zsigrai [a,*], Tam Cong Nguyen [b], Andrey Berlizov [a,1,2]

[a] European Commission, Joint Research Centre (JRC), Institute for Transuranium Elements (ITU), 76125 Karlsruhe, P.O. Box 2340, Germany
[b] Centre for Energy Research of the Hungarian Academy of Sciences (EK), 1525 Budapest 114, P.O. Box 49, Hungary

ARTICLE INFO

Article history:
Received 18 November 2014
Received in revised form 30 June 2015
Accepted 7 July 2015
Available online 4 August 2015

Keywords:
^{232}U
Gamma spectrometry
Reprocessed uranium
Nuclear safeguards
Nuclear forensics

ABSTRACT

The ^{232}U content of various uranium-bearing items was measured using low-background gamma spectrometry. The method is independent of the measurement geometry, sample form and chemical composition. Since ^{232}U is an artificially produced isotope, it carries information about previous irradiation of the material, which is relevant for nuclear forensics, nuclear safeguards and for nuclear reactor operations. A correlation between the ^{232}U content and ^{235}U enrichment of the investigated samples has been established, which is consistent with theoretical predictions. It is also shown how the correlation of the mass ratio ^{232}U/^{235}U vs. ^{235}U content can be used to distinguish materials contaminated with reprocessed uranium from materials made of reprocessed uranium.

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1. Introduction

In this work a non-destructive method is presented for measuring the ^{232}U content of uranium-bearing items by gamma spectrometry. The original aim was to extend the nuclear forensics toolbox helping to trace the origin and history of illicit nuclear material. However, the method can also be applied in other fields where the knowledge of the ^{232}U content is relevant, such as nuclear reactor operation, nuclear safeguards and nuclear arms control.

For nuclear forensics it is important that ^{232}U typically does not occur in natural uranium in measurable quantities, but is formed during the irradiation of uranium or thorium in a nuclear reactor. Therefore, if ^{232}U is found then it carries information about the history of the material in which it was detected. If ^{232}U is present, then it means that the sample contains some irradiated material (e.g. reprocessed uranium) or is contaminated with such [1]. This information helps to trace the origin of illicit nuclear material [2].

For nuclear-reactor operators the presence of ^{232}U in uranium fuel is relevant because it implicitly implies the presence of ^{236}U, which is a neutron absorber and influences the operation of a nuclear reactor (see, e.g. page 11 in [3]). While the direct measurement of ^{236}U in reactor fuel is only possible by destructive methods, ^{232}U in reactor fuel can be measured non-destructively by gamma spectrometry [4].

The detection of the isotope ^{232}U had also been proposed for confirming the presence and distribution of highly enriched uranium (HEU) in nuclear weapons [5]. In [6] it has been argued that the presence of ^{232}U can be an unclassified attribute of HEU for nuclear arms control. Furthermore, ideas on using small amounts of ^{232}U added to uranium have been proposed in [7] and [8] to help prevent the proliferation of nuclear weapons.

Two standards (see [9]), ASTM C 996 [10] and ASTM C 787 [11] define the limits on the ^{232}U and ^{236}U contents of the feed to an enrichment process and of uranium enriched to less than 5% ^{235}U. For natural U the limit is defined relative to total U, while for enriched U it is defined relative to ^{235}U, as follows:

- "Commercial natural uranium" (CNU) [10]:
  - ^{232}U content $<1 \times 10^{-11}$ g/g U and ^{236}U content $<2 \times 10^{-5}$ g/g U.
- "Enriched commercial grade uranium" (ECGU) [11]:
  - ^{232}U content $<2 \times 10^{-9}$ g/g ^{235}U and ^{236}U content $<5 \times 10^{-3}$ g/g ^{235}U.

Upon enrichment to 5% ^{235}U "Commercial natural uranium" (CNU) will become "Enriched commercial grade uranium"

* Corresponding author.
[1] Present address: International Atomic Energy Agency, Vienna International Centre, PO Box 100, 1400 Vienna, Austria.
[2] Former address: Institute for Nuclear Research, Prospekt Nauky, 47, 03680 Kyiv, Ukraine.

http://dx.doi.org/10.1016/j.nimb.2015.07.047
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(ECGU) which satisfies the specifications of ASTM-C996. Lower ^{232}U and ^{236}U contents are considered to be due to trace contamination by irradiated uranium and, from the point of view of transport, storage and handling, the material is treated as unirradiated uranium of natural origin.

The method described in this paper can show whether a sample satisfies the criteria of ASTM C787 and ASTM C996 for CNU and ECGU. To support nuclear-forensic investigations, even minor traces of reprocessed uranium in ECGU can be detected proving if a sample originates from a facility which handles reprocessed uranium.

The method is applicable to a wide range of samples, from the smallest ones containing less than 1 g of uranium, to complete nuclear-reactor fuel assemblies. The measurable ^{232}U content varies in a range of 4 orders of magnitude. If the sample is homogenous, then the ^{232}U content obtained by the described method is independent of the measurement geometry, sample form and chemical composition.

2. Theoretical prediction of the ^{232}U content of Uranium

The artificial nuclide ^{232}U can form in a variety of nuclear reaction chains. The most important were given, for example, in [6,12–15]. These reactions occur, e.g., during the irradiation of uranium or thorium fuel in a nuclear reactor.

To estimate the ^{232}U content of spent nuclear fuel we used the webKORIGEN depletion calculation engine available within Nucleonica [16]. We calculated the approximate ^{232}U content remaining in spent reactor fuel 6 years after the end of irradiation for fuel of 4% initial ^{235}U enrichment.[3] For spent pressurized water reactor fuel for a range of burn-ups between 15 and 60 MWd/kg U we calculated that there is 3.82×10^{-8} to 5.82×10^{-7} mass% of ^{232}U relative to total U. We also estimated that the ^{232}U content relative to the remaining ^{235}U is in the range from 1.48×10^{-8} to 1.40×10^{-6} g/g ^{235}U. These values are in accordance with previous estimates reported in [14,15].

In [13] the ^{232}U content of uranium was estimated from burn-up calculations and from simple mathematical models of the enrichment cascades. It was concluded in [13] that depleted uranium contains 1600–8000 times less ^{232}U than HEU. Furthermore, it has been estimated in [13] that cascade enrichment increases the ^{232}U concentration by a factor of 200–1000.

When uranium from reprocessed spent fuel is used to make new fuel for nuclear reactors, it is usually blended with other uranium materials to adjust the ^{235}U enrichment of the product to a specified value. Therefore, the final ^{232}U content of the product is less than that of the spent fuel.

3. Instruments and materials

For the studies presented in this paper spectra were taken with 4 different HPGe detectors at 4 different locations (see Table 1).

The majority of the spectra were taken by a low-background HPGe detector (EK1) located at the Department for Nuclear Security of the Centre for Energy Research in Budapest, Hungary. Spectra of research-reactor fuel rods were taken with a detector (EK2) on site of the research reactor of the Centre for Energy Research. Spectra of certified reference materials were also taken by a detector at the Institute of Nuclear Research in Kyev, Ukraine (INR). Finally, various spectra were taken at the Institute of Transuranium Elements of the Joint Research Centre of the European Commission in Karlsruhe, Germany (detectors ITU1 and ITU2). Table 1 summarizes the detectors used.

Table 1
Gamma spectrometers used in this work.

Detector short name	Location	Manufacturer	Measured FWHM at 1332 keV (kev)	Declared efficiency (%)*
EK1	Budapest, Hungary	PGT	2.05	34
EK2	Budapest, Hungary	Canberra	1.82	35
INR	Kyiv, Ukraine	Canberra	1.78	63
ITU1	Karlsruhe, Germany	Canberra	1.86	54
ITU2	Karlsruhe, Germany	Ortec	1.78	52

* In the standard definition, at 1332 keV and 25 cm source-detector distance, relative to a 3″ × 3″ NaI(Tl) detector.

The items investigated in this work are listed in Table 5 in the Appendix, together with some basic information about them. The samples included, among others, certified reference materials, seized fuel pellets, research-reactor fuel rods and U metal, spanning an enrichment range from depleted to highly enriched uranium.

4. The method for measuring the ^{232}U content of uranium by gamma spectrometry

4.1. General description of the method

In most cases the gamma radiation coming directly from ^{232}U cannot be detected by gamma spectrometry because the ^{232}U gamma peaks are masked by the Compton background of the peaks from the major uranium isotopes. However, the daughter products of ^{232}U, in particular ^{212}Pb, ^{212}Bi, and ^{208}Tl emit strong gamma radiation detectable by gamma spectrometry. All these three isotopes are short-lived and they are in equilibrium with ^{228}Th.

These isotopes, however, are also present in the decay chain of ^{232}Th (see Fig. 1), and the presence of the gamma-emitting nuclides ^{212}Pb, ^{212}Bi, and ^{208}Tl might be also due to the presence of ^{232}Th in the sample. The two decay chains merge at ^{228}Th. Therefore, the activity of ^{228}Th, as well as of its short-lived gamma-emitting daughters ^{212}Pb, ^{212}Bi and ^{208}Tl, can be given as the sum of two terms: one of them accounting for the build-up from ^{232}Th and another accounting for the build-up from ^{232}U. This is reflected in the following equation, which can be obtained using the Bateman solution to the equations of radioactive decay [17]:

$$\begin{aligned}\frac{A_{\mathrm{Tl208}}}{p} &= A_{\mathrm{Bi212}} = A_{\mathrm{Pb212}} = A_{\mathrm{Th228}} \\ &= A_{\mathrm{Th232}}\left[1 + \frac{\lambda_{\mathrm{Th228}}\exp(-\lambda_{\mathrm{Ra228}}t) - \lambda_{\mathrm{Ra228}}\exp(-\lambda_{\mathrm{Th228}}t)}{\lambda_{\mathrm{Ra228}} - \lambda_{\mathrm{Th228}}}\right] \\ &\quad + A_{\mathrm{U232}}\left[\frac{1 - \exp((\lambda_{\mathrm{U232}} - \lambda_{\mathrm{Th228}})t)}{1 - \lambda_{\mathrm{U232}}/\lambda_{\mathrm{Th228}}}\right] \qquad (1)\end{aligned}$$

where $p = 0.359$ [16] is the decay branching probability of the decay of ^{212}Bi to ^{208}Tl (see Fig. 1), λ_{Th228}, λ_{Ra228} and λ_{Th232} are the respective decay constants, while A_{Tl208}, A_{Bi212}, A_{Pb212}, A_{Th228}, A_{Th232} and A_{U232} are the corresponding activities at the time of the measurement. Note that Eq. (1) was obtained taking into account that $\lambda_{\mathrm{Th228}} \gg \lambda_{\mathrm{Th232}}$ and $\lambda_{\mathrm{Ra228}} \gg \lambda_{\mathrm{Th232}}$.

[3] The webKORIGEN settings used for the calculations were the following: Mode of calculation: reactor irradiation and decay; reactor type: PWR; fuel: uranium oxide with 4.0% enrichment; cross section library: "PWR UOX 4.0% U235 60 MWd/kgHM"; length of cycle: 1 y; Number of cycles: 2 for 15 MWd/kg U and 5 for 50–60 MWd/kg U; load factor: 80.0%; fuel decay time after discharge: 6 y, heavy metal mass: 20 t.

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