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Isotopic “fingerprints” for natural uranium ore samples

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Abstract

A set of six samples, collected worldwide from various uranium ore mining facilities, was analysed for uranium isotopic composition by high accuracy isotope mass spectrometry. The goal of this article was twofold: to measure isotopic variations between samples of different geographical origin and to produce calibrated isotope ratios with the smallest achievable uncertainty (as defined according to the ISO Guide to the Expression of Uncertainty in Measurement). In the first step, the molar ratio of the isotopes ^{235}U and ^{238}U, $n(^{235}\text{U})/n(^{238}\text{U})$, was measured using a UF_6-gas-inlet isotope mass spectrometer (VARIAN MAT 511). This instrument was calibrated against gravimetrically prepared synthetic isotope mixtures thus allowing SI-traceable measurements to be made. The ratios of the “minor isotopes” to ^{238}U [$n(^{234}\text{U})/n(^{238}\text{U})$ and $n(^{236}\text{U})/n(^{238}\text{U})$] were determined in a second step using a thermal ionisation mass spectrometer with high abundance sensitivity (Finnigan MAT262-RPQ-PLUS). The mass-fractionation correction was done internally using the result of the $n(^{235}\text{U})/n(^{238}\text{U})$ measurement. As a result, the complete measured uranium isotopic composition is traceable to the SI system. For all ratios $n(^{234}\text{U})/n(^{238}\text{U})$, $n(^{235}\text{U})/n(^{238}\text{U})$, and $n(^{236}\text{U})/n(^{238}\text{U})$ significant differences for samples of different origin were found. Regarding the $n(^{236}\text{U})/n(^{238}\text{U})$ results, only two samples, one of them from the Oklo reactor in Gabon, showed significant presence of ^{236}U. For all other samples an upper limit for $n(^{236}\text{U})/n(^{238}\text{U})$ of about 6×10^{-10}, mainly dependent on the instrumentation, was found. As a result of this study we propose values for the isotope abundances of natural uranium for the “Best Measurement from a Single Terrestrial Source” and the “Range of Natural Variations” in the IUPAC-table of the “Isotopic Composition of the Elements.” (Int J Mass Spectrom 193 (1999) 9–14) © 1999 Elsevier Science B.V.

Keywords: Natural uranium; Abundance sensitivity; IUPAC table

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1. Introduction

Uranium, the heaviest naturally occurring element on earth, plays an important role in daily life because of its use in nuclear power plants. Release of nuclear waste into the environment is an important issue in many parts of the world. Anthropogenic uranium can be found in a higher concentration compared with natural uranium ($\cong 6\ \mu$g/g) and typically does not have the same isotopic composition. There are three naturally occurring uranium isotopes with long half-lifes, ^{234}U, ^{235}U, and ^{238}U, the isotope abundance ratios being approximately 0.000055:0.00725:1. These ratios may have small variations depending on the geographic origin of the sample, due to natural isotope fractionation, nuclear reactions or anthropogenic contamination. Variations of the ratio $n(^{234}\text{U})/n(^{238}\text{U})$ may also arise from processes that disturb the secular equilibrium between these isotopes.

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Anthropogenic uranium also contains small amounts of ^{236}U, formed by neutron capture of ^{235}U in nuclear industrial processes, the ratio $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ranging between 10^{-7} and 10^{-2} [1]. Some ^{236}U has only been found in exceptional geological specimens (e.g. samples from the Oklo reactor). As the natural $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ abundance ratio is expected to be extremely low, arising from activation processes as cosmic ray activation, mass spectrometric instrumentation that is able to measure isotope abundance ratios within a dynamic range of 8–10 orders of magnitude is required.

Because the lowest abundant isotope ^{236}U is close in mass to ^{238}U, the most abundant one, the influence of the ^{238}U ion beam on that of ^{236}U from the ^{238}U peak tail can be observed to some extent in every isotope ratio mass spectrometer. The property used to describe this influence is called the "abundance sensitivity." The value of the abundance sensitivity at ^{236}U arising from the tail of ^{238}U ("$^{238}\mathrm{U} - 2$ u") is defined by the ratio $I^{(236)}/I^{238}$, where I^{236} is the ion current of ^{238}U at mass 236 and I^{238} that at mass 238. Usually the abundance sensitivity of a mass spectrometer is given one mass unit below the tailing peak ("$^{238}\mathrm{U} - 1$ u"), but in this context its value 2 mass units below (at ^{236}U) is the critical one. Commercial quadrupole or single-focusing magnetic sector mass spectrometers usually have an abundance sensitivity in the range of 10^{-5}–10^{-7} at "$^{238}\mathrm{U} - 1$ u," leading to about 10^{-6}–10^{-8} at "$^{238}\mathrm{U} - 2$ u," which is the limiting factor for the detection and measurement of ^{236}U.

Most of the values published in literature do not have well-defined uncertainty statements. The approach used here is based on the ISO Guide on the Expression of Uncertainty in Measurement [2] and the values given here are therefore not merely repeatability or reproducibility values only. For the investigation of differences between samples, repeatability alone is sufficient. Values better than 0.01% repeatability in the literature for uranium isotope ratios can be found. On the other hand, full uncertainties according to the ISO Guide which are better than 0.03% have rarely been published (e.g. by Rosman et al. [3]). In this work, the objective was also to produce calibrated ratios with the smallest combined uncertainties for these samples. By doing this, values in the IUPAC Isotopic Table [4] could be based on data of the highest metrological quality. For this reason a combination of two highly accurate mass spectrometric techniques was used. The $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ ratio was measured with an uncertainty of 0.05% (coverage factor $k = 2$, according to the ISO Guide [2]) using a $\mathrm{UF_6}$-gas-inlet isotope mass spectrometer, which was calibrated using gravimetrically prepared synthetic isotope mixtures.

The isotope ratios of the "minor" isotopes relative to ^{238}U, $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ and $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$, are determined by thermal ionisation mass spectrometry with a "high" abundance sensitivity of better than 1.2×10^{-10} at "$^{238}\mathrm{U} - 2$ u" (Finnigan MAT262-RPQ-PLUS, [5]), using the result of $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ obtained by $\mathrm{UF_6}$-gas-inlet isotope mass spectrometry for internal mass fractionation correction. Therefore, the results for $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ and $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ obtained by thermal ionisation mass spectrometry can also be considered as "calibrated" results and the complete uranium isotopic composition measured in this way is traceable to the SI system.

The effort done to produce calibrated values for uranium isotope ratios with reliable uncertainties serves also one other important goal: to improve reliability and comparability in environmental nuclear safeguards, especially for the detection of possible undeclared nuclear activities. A significant depletion of the isotope amount ratio $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ compared to the "normal terrestrial value" is an indication of nuclear fission processes having occurred in the sample. Since the consequences of any possible (undeclared) nuclear activity are severe, conclusions of such mass spectrometric analyses have to be based on high quality data for uranium isotope ratios, having the best possible reliability and being traceable to a common system of units, i.e. the SI system.

The ratio $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ can also be used to detect nuclear activities as the ^{236}U is formed by a neutron capture process. As this ratio is very small, the dominant source of uncertainty arising from the reproducibility of the measurement, the calibration will only be a minor contributor to the total uncertainty.

Table 1
Origin of uranium ore samples analysed in this study and in [1]

No.	Country of origin	Milling facility
1	Gabon	Comuf Mounana
2	Czech Republic	DIAMO, Straz pod Ralskem
3	Canada	CAMECO Rabbit Lake Op.
4	Namibia	Roessing Uranium Mine
5	France	Cogema Lodeve
6	France	CETAMA Amethyste
[1], 1	Finland	Askola
[1], 2	Finland	Paukkajanvaara
[1], 3	Australia	Ranger Mine
[1], 4	Australia	Dam Operations
[1], 5	Canada	Cogema Resources
[1], 6	Canada	CAMECO Key Lake Op.

This study constitutes a continuation of a previous work on natural uranium samples by Ovaskainen et al. [1]. Because of the improved abundance sensitivity (factor ≅ 8) and therefore improved detection limit for ^{236}U of the Finnigan MAT262 RPQ-PLUS compared with the RPQ-Prototype used in [1], some of the samples were reanalysed and some more uranium ore samples were included. The origin for all samples is shown in Table 1.

2. Experimental

2.1. Chemical preparation and purification of the uranium ore samples

The mass spectrometric determination of uranium with high precision and accuracy requires careful chemical purification procedure prior to mass spectrometric measurements. In this work the samples were split into two subsamples: subsample 1 for conversion into UF_6 for gas-inlet isotope mass spectrometry and subsample 2 for conversion to $UO_2(NO_3)_2$ and further purification using ion exchange columns [1]. The final 1 M HNO_3 solution of subsample 2 had a uranium concentration of 5 mg/ml. A 2 μL (10 μg) uranium sample was deposited on the centre of the rhenium filament of a double filament source for measurement by thermal ionisation mass spectrometry.

2.2. Mass spectrometric measurements

2.2.1. Determination of $n(^{235}U)/n(^{238}U)$ by UF_6-gas-inlet isotope mass spectrometry

The measurements were performed using the double standard set IRMM-EC-171 consisting of two gravimetrically prepared isotope reference materials. The measurements were calibrated against these two isotopic reference materials, one of them having a slightly higher and the other one having a slightly lower $n(^{235}U)/n(^{238}U)$ ratio compared to the sample.

2.2.2. Determination of $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ by thermal ionisation mass spectrometry

A Finnigan MAT262-RPQ-PLUS thermal ionisation mass spectrometer was used to measure the minor isotope ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$. In order to avoid any problems concerning Faraday cup intercalibration, the multicollector array was not used. The so-called minor isotopes ^{234}U and ^{236}U can only be detected using a secondary electron multiplier (SEM), which is used in combination with a deceleration lens system to improve the abundance sensitivity, the “RPQ-PLUS.” The working principle of the RPQ-PLUS lens system is explained in detail in [5] and [6].

With the aid of the RPQ-PLUS deceleration lens system no significant tail contribution at mass 236 could be detected. The measurement of the tail contribution was limited by the background rate $0.62 \pm 0.29\ \text{min}^{-1}$, which was determined by counting at mass 236 for 30 min, using unloaded filaments at the usual run temperatures. The background rate already includes the dark noise $0.114 \pm 0.065\ \text{min}^{-1}$ of the SEM. The higher value of the background rate can be explained by a possible ^{236}U contamination in the ion source or scattering of ions in the analyser tube, e.g. Re^+, Na^+, or K^+ ions, which are always emitted from the filaments.

The instrumental detection limit for the ratio $n(^{236}U)/n(^{238}U)$ can be calculated in the conventionally defined way as three times the standard uncertainty of the background rate, divided by the typical ^{238}U intensity ($^{238}U \cong 2 \times 10^{-11}$ A). This yields a value for the detection limit of 1.2×10^{-10}. If the SEM dark noise alone were considered, the detection limit for the ratio $n(^{236}U)/n(^{238}U)$ would be 2.6×10^{-11}.

The relative yield of the RPQ-PLUS ion counter versus a Faraday cup (≅98%) is measured in a separate procedure before each run by switching the ^{235}U beam between the RPQ ion counter and the Faraday cup. This procedure is usually done at a ^{235}U ion beam intensity of about 300000 counts/s, corresponding to an ion current of about 5×10^{-14} A. The relative yield of the RPQ-PLUS measured in this way includes both the relative yield of the ion counter (>98%) and the transmission of the RPQ lens system (≅100%, [5]), as these properties can not be measured separately. It has been shown that over a 2 h period the standard uncertainty of the RPQ-PLUS yield is about ±0.2%. This value can be explained from the ion counting statistics and the uncertainties due to the Faraday cup baseline noise and the Faraday cup signal noise in the calibration procedure. As a consequence, the two ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ cannot be measured with a standard uncertainty better than ±0.2% with the present experimental method.

For each of the six samples, six replicate filament loadings were prepared and measured. The repeatability was compared with the "combined uncertainty" of a single measurement, which was calculated by propagating the contributions of all uncertainty sources involved. As explained in detail for sample no. 2 in [7], the maximum of the two can be considered as a realistic final value of the uncertainty. An expanded uncertainty, calculated using a coverage factor $k = 2$, was applied to all uncertainties of isotope amount ratios given in this study.

3. Results

For all the ratios, $n(^{234}U)/n(^{238}U)$, $n(^{235}U)/n(^{238}U)$, and $n(^{236}U)/n(^{238}U)$ some significant differences for samples of different geographical origin were found. The measured isotope ratios are given in Table 2.

Table 2
Isotope ratios $n(^{234}U)/n(^{238}U)$, $n(^{235}U)/n(^{238}U)$, and $n(^{236}U)/n(^{238}U)$, measured for six uranium ore samples[a]

No.	$n(^{234}U)/n(^{238}U)$	$n(^{235}U)/n(^{238}U)$	$n(^{236}U)/n(^{238}U)$
1	$5.434(42) \times 10^{-5}$	0.007 235 7(36)	$9.32(69) \times 10^{-9}$
2	$8.355(49) \times 10^{-5}$	0.007 256 0(36)	$2.540(92) \times 10^{-8}$
3	$5.444(48) \times 10^{-5}$	0.007 256 8(36)	$2.8(1.7) \times 10^{-10}$
4	$5.460(41) \times 10^{-5}$	0.007 256 8(36)	$2.0(1.8) \times 10^{-10}$
5	$5.154(28) \times 10^{-5}$	0.007 257 2(36)	$1.9(1.7) \times 10^{-10}$
6	$5.340(33) \times 10^{-5}$	0.007 235 2(36)	$2.0(1.4) \times 10^{-10}$

[a] Uncertainties calculated according to the ISO Guide to the Expression of Uncertainty in Measurement [2], using a coverage factor $k = 2$.

For $n(^{234}U)/n(^{238}U)$, the results of nos. 2, 5, and 6 differ significantly from the "natural" isotope amount ratio of $5.499(18) \times 10^{-5}$, calculated from the half-lifes of these isotopes [8]. The $n(^{234}U)/n(^{238}U)$ ratio of no. 2, coming from the Czech Republic (see Table 1 for origin of samples), is very exceptional because it is much higher than the calculated equilibrium value. This cannot be explained by geological processes; one possibility may be because of some anthropogenic contamination with Pu, especially ^{238}Pu.

The $n(^{235}U)/n(^{238}U)$ ratio in sample no. 1 (from the natural reactor near Oklo, Gabon) as well as in no. 6 are slightly depleted, compared to the well-known "normal terrestrial" ratio of about 0.00725. The "original" $n(^{235}U)/n(^{238}U)$ ratio was established in the r process (rapid neutron capture) of nucleosynthesis in a stellar, presolar environment and has then decreased due to α decay since that time, leading to the present terrestrial ratio.

A significant presence of ^{236}U was only found for nos. 1 and 2, indicating to the previous existence of neutron processes in these samples. Applying a coverage factor of 3, for all other samples an upper limit for $n(^{236}U)/n(^{238}U)$ of about 6×10^{-10} was found. Their $n(^{236}U)/n(^{238}U)$ ratio—and consequently also their ^{236}U content—is therefore very close to the background level.

4. Conclusions

Except for the samples nos. 3 and 4 there is no pair of two samples in the set of six, which have the same isotopic composition. So, with some constraints, this set of data for the six natural uranium ore samples can be considered as isotopic “fingerprints” for their geographic origin. When looking only at one isotope ratio, it is impossible to distinguish between most of the samples. Concerning their $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ isotope ratios, the possibility of distinguishing between different samples is still limited by the instrumental detection limit. The natural abundance of $^{236}\mathrm{U}$ can still be much lower than that detectable by the present instrument. However, experiments using accelerator mass spectrometry methods of measurement have shown a similar detection limit [9].

Table 3
“Best measurement from a single terrestrial source”, present IUPAC values compared to suggestion for replacement

	Present IUPAC value, 1s, N*[a] (at. %)	Suggested value (no. 4), $k = 2$, C*[b] (at. %)
$^{234}\mathrm{U}$	0.005 48(2)	0.005 420(42)
$^{235}\mathrm{U}$	0.720 0(1)	0.720 41(36)
$^{238}\mathrm{U}$	99.274 5(10)	99.274 17(36)

[a] *N: without use of calibrated mixtures for mass fractionation correction: the abundance of $^{234}\mathrm{U}$ was not measured for the same sample as the abundances of $^{235}\mathrm{U}$ and $^{238}\mathrm{U}$.

[b] *C: measurement using calibrated mixtures, SI-traceable results. The uncertainties were calculated according to the ISO Guide to the Expression of Uncertainty in Measurement [2], using a coverage factor $k = 2$.

5. Suggestions for the “Best Measurement from a Single Terrestrial Source” and “Range of Natural Variations” in the IUPAC table

In the latest IUPAC table of the “Isotopic Composition of the Elements 1997” [4] a set of noncalibrated results for isotope abundances of natural uranium is considered as the Best Measurement from a Single Terrestrial Source. This result for $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$, published by Cowan and Adler in 1976 [10], was not calibrated using gravimetrically prepared isotope mixtures. Furthermore for the $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ tabulated value, published by Smith and Jackson in 1969 [11], the $^{234}\mathrm{U}$ abundance was measured relative to a $^{233}\mathrm{U}$ spike, but it was neither corrected for mass fractionation nor related to the value for $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ given by Cowan and Adler [10]. In contrast, within this new study all $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ ratios have been calibrated using synthetic isotope mixtures. The $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ ratio was measured within the same run and corrected for mass fractionation using the $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ ratio as internal standard. We therefore propose that these new results, traceable to the SI system, should be considered to replace the present data in the IUPAC table [4]. For the comparison from the method point of view with previous results the sample amount has also to be taken into account. Calibration using synthetic isotope mixtures usually requires a larger amount of sample, which is not always available for normal terrestrial material.

The isotopes $^{233}\mathrm{U}$ and $^{236}\mathrm{U}$ are considered as unstable (half-life $< 4 \times 10^8$a) and are therefore not mentioned in the IUPAC table [4].

Regarding the six individual samples analysed in this study, no. 1 from the Oklo reactor is a “geologically exceptional specimen” (terminology used in [4]). No. 2 is exceptional in the $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ ratio most likely because of an anthropogenic contamination. Nos. 5 and 6 are depleted in $^{234}\mathrm{U}$, which can be explained geologically by preferential leaching of $^{234}\mathrm{U}$ from the ore bodies [1].

From the pair of the remaining samples (nos. 3 and 4) the result of no. 4 is proposed as the Best measurement from a Single Terrestrial Source, because the $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ ratio has a smaller uncertainty and also a lower limit was found for the $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ratio. In Table 3 the abundances of the naturally occurring isotopes $^{234}\mathrm{U}$, $^{235}\mathrm{U}$, and $^{238}\mathrm{U}$, measured for sample no. 4, are compared with the present values in the IUPAC table [4]. In this context, it is vital to stress that true combined uncertainties are given throughout in this article, in accordance with the ISO Guide on the Expression of Uncertainty in Measurement [2], and not repeatabilities nor reproducibilities as in the majority of published data.

In addition, a Range of Natural Variations is also given in the IUPAC table [4]. Samples nos. 3, 4, 5, and 6 of this study together with six different samples of natural uranium investigated in [1] are tabulated together in Table 4. A suggestion for the Range of

Table 4
Isotope abundance ratios $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$, $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$, and $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ of natural uranium samples from this study (nos. 3, 4, 5 and 6) and from [1][a]

No.	$n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$	$n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$	$n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$
3	$5.444(48) \times 10^{-5}$	0.007 256 8(36)	$2.8(1.7) \times 10^{-10}$
4	$5.460(41) \times 10^{-5}$	0.007 256 8(36)	$2.0(1.8) \times 10^{-10}$
5	$5.154(28) \times 10^{-5}$	0.007 257 2(36)	$1.9(1.7) \times 10^{-10}$
6	$5.340(33) \times 10^{-5}$	0.007 235 2(36)	$2.0(1.4) \times 10^{-10}$
[1], 1	$5.444(08) \times 10^{-5}$	0.007 254 2(36)	2.16×10^{-8}
[1], 2	$5.126(76) \times 10^{-5}$	0.007 257 8(36)	1.45×10^{-8}
[1], 3	$5.455(44) \times 10^{-5}$	0.007 258 1(36)	$<1.45 \times 10^{-8}$
[1], 4	$5.341(62) \times 10^{-5}$	0.007 258 4(36)	$<1.45 \times 10^{-8}$
[1], 5	$5.385(60) \times 10^{-5}$	0.007 259 6(36)	$<1.45 \times 10^{-8}$
[1], 6	$5.397(34) \times 10^{-5}$	0.007 260 3(36)	$<1.45 \times 10^{-8}$

[a] Uncertainties were calculated according to the ISO Guide to the Expression of Uncertainty in Measurement [2], using a coverage factor $k = 2$.

Natural Variations, based on the values in Table 3, is given in Table 5. Sample no. 6 of this study is remarkable because of its strong depletion in $^{235}\mathrm{U}$, more than three times the entire range of all the other samples, its depletion is similar to sample no. 1 from the Oklo natural reactor. We believe, however, that this particular sample is not a natural sample in the accepted sense, but contains anthropogenic material or possibly material from the Oklo reactor and for this reason we have not included it in the Range of Natural Variations.

It seems that anthropogenic contamination has to be taken into account increasingly in such analyses, which makes calibrated measurements and comparability even more important. Since all results taken into account are based on calibrated measurements the proposed calibrated range for natural variations is linked to the calibrated measurement suggested for the Best Measurement from a Single Terrestrial Source.

Table 5
Suggestion for the “Range of natural variations” in the IUPAC table[a]

	Present IUPAC range of natural variations (at. %)	Suggested range of natural variations (at. %)
$^{234}\mathrm{U}$	0.0059–0.0050	0.0054–0.0051
$^{235}\mathrm{U}$	0.7202–0.7198	0.7207–0.7201
$^{238}\mathrm{U}$	99.2752–99.2739	99.2748–99.2739

[a] The suggested natural range is derived from sample nos. 3, 4, 5 of this study together with the data of six samples measured previously [1]. All results taken into account for the suggested new range are based on calibrated measurements, their uncertainties were calculated according to the ISO Guide to the Expression of Uncertainty in Measurement [2], using a coverage factor $k = 2$.

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