



Standard Test Method for Isotopic Abundance Analysis of Uranium Hexafluoride and Uranyl Nitrate Solutions by Multi-Collector, Inductively Coupled Plasma-Mass Spectrometry¹

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1. Scope

1.1 This test method covers the isotopic abundance analysis of ^{234}U , ^{235}U , ^{236}U and ^{238}U in samples of hydrolysed uranium hexafluoride (UF_6) by inductively coupled plasma source, multicollector, mass spectrometry (ICP-MC-MS). The method applies to material with ^{235}U abundance in the range of 0.2 to 6 % mass. This test method is also described in ASTM STP 1344.

1.2 The values stated in SI units are to be regarded as standard. No other units of measurement are included in this standard.

1.3 *This standard does not purport to address all of the safety concerns, if any, associated with its use. It is the responsibility of the user of this standard to establish appropriate safety and health practices and determine the applicability of regulatory limitations prior to use.*

2. Referenced Documents

2.1 ASTM Standards:²

C 761 Test Methods for Chemical, Mass Spectrometric, Spectrochemical, Nuclear, and Radiochemical Analysis of Uranium Hexafluoride

C 787 Specification for Uranium Hexafluoride for Enrichment

C 996 Specification for Uranium Hexafluoride Enriched to Less Than 5 % ^{235}U

D 1193 Specification for Reagent Water

2.2 Other Document:

STP 1344 Applications of Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) to Radionuclide Determinations³

¹ This test method is under the jurisdiction of ASTM Committee C26 on Nuclear Fuel Cycle and is the direct responsibility of Subcommittee C26.05 on Methods of Test.

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² For referenced ASTM standards, visit the ASTM website, www.astm.org, or contact ASTM Customer Service at service@astm.org. For *Annual Book of ASTM Standards* volume information, refer to the standard's Document Summary page on the ASTM website.

³ Available from ASTM Headquarters.

3. Terminology

3.1 Acronyms:

3.1.1 *amu*—atomic mass unit

3.1.2 *ICP-MC-MS*—Inductively Coupled Plasma Multi-Collector Mass Spectrometer

3.1.3 *ICP-MS*—Inductively Coupled Plasma Mass Spectrometer

3.1.4 *UIRM*—Uranium Isotopic Reference Material

4. Summary of Test Method

4.1 Samples are received either in the form of uranium hexafluoride (UF_6) or aqueous uranic solution. The UF_6 samples are hydrolysed, diluted and acidified with nitric acid. Uranic solution samples are diluted and acidified with nitric acid. If required, an internal reference of thorium isotopes can be subsequently added to each diluted sample. As detailed in Section 8, isotope pairs of elements other than thorium could be used for an internal reference.

4.2 The samples are contained in polypropylene tubes that are inserted into the auto-sampler rack of the mass spectrometer. Sample details are input to the computer and the instrument is prepared for measurement. The automatic measuring sequence is initiated.

4.3 Uranium Isotopic Reference Materials (UIRMs) are used to calibrate the instrument. Each UIRM is prepared in aqueous solution (acidified with nitric acid) and if required spiked with the same internal reference as the samples. This calibration solution is measured and a mass bias parameter is calculated that is stored and subsequently imported into each of the sample measurements⁴ to correct the measured uranium isotopic ratios.

4.4 Measurements of isotopic ratios in the calibration solution and the subsequent samples are initiated by customised software. The mass bias factor is computed from the measured isotopic ratios in the calibration solution. This parameter is then exported to correct the measured isotopic ratios of the samples for mass bias. The corrected isotopic abundances are

⁴ The uranium isotopic precision of measurement, limit of detection and uncertainty of measurement are listed in Section 15 and Appendix X1.

expressed as % atomic and are converted to % mass prior to reporting. Details of the mass bias correction are presented in [Appendix X1](#).

5. Significance and Use

5.1 The test method is capable of measuring uranium isotopic abundances of ^{234}U , ^{235}U , ^{236}U and ^{238}U as required by Specifications [C 787](#) and [C 996](#).

6. Interferences

6.1 *Mass Bias*—Electrostatic repulsion between uranium ions causes a so-called “mass bias” effect. Mass bias is observed as an enhancement in the number of ions detected at the collectors from the heavier uranium isotopes relative to the lighter uranium isotopes. A calibration procedure is used to correct the mass spectrometer for mass bias.

6.2 *Adjacent Isotopic Peaks*—The abundance sensitivity of the ICP-MC-MS at mass 237 is specified to be less than 0.5 parts per million of the ^{238}U ion beam. The method is limited to the measurement of ^{235}U isotopic abundances below 6 %, consequently interference effects with the ^{234}U and ^{236}U ion beams are negligible.

6.3 *Isobaric Molecular Interferences*—A molecular interference exists at mass 236 between ^{236}U and a hydride of ^{235}U , which is formed in the plasma. This interference can be corrected by measuring the beam height of the ^{238}U hydride at mass 239, and applying the correction defined in Eq 1, to the measured ^{236}U ion beam:⁵

$$^{236}\text{U}_c = ^{236}\text{U}_m - \left(^{235}\text{U} \times \frac{^{238}\text{UH}}{^{238}\text{U}} \right) \quad (1)$$

where:

- $^{236}\text{U}_c$ = the corrected ^{236}U ion beam,
- $^{236}\text{U}_m$ = the measured ^{236}U ion beam,
- ^{235}U = the measured ^{235}U ion beam,
- ^{238}UH = the measured ^{238}U hydride ion beam, and
- ^{238}U = the measured ^{238}U ion beam.

6.4 Memory Effects:

6.4.1 Contamination of the sample introduction system from previous samples produces memory interference effects. Such effects are accentuated when samples that are depleted in ^{235}U are measured after enriched samples. Memory effects can be readily assessed by aspirating a 0.3 M nitric acid solution and measuring the background ^{238}U ion beam. The sample introduction system should be periodically disassembled and cleaned, to minimise the background ^{238}U ion beam.

6.4.2 A background correction is performed during the measurement run by monitoring the analyte signals of the 0.3 M nitric acid rinse solution. The background correction is measured prior to the mass calibration and is re-measured before each subsequent sample.

7. Apparatus

7.1 Mass Spectrometer:

7.1.1 The mass spectrometer has an inductively coupled plasma (ICP) source and a double focusing electrostatic/magnetic sector analyser equipped with twelve Faraday detectors and two ion counters.⁶

7.1.2 The mass spectrometer is fully computer controlled using customised software and is equipped with an auto-sampler.

7.2 *Polypropylene Sample Tubes, Screw-Cap*, 50 mL.

7.3 *Polypropylene Sample Tubes, Screw-Cap*, 10 mL.

7.4 *Positive Displacement Pipette, and Tips to Suit*, 0.01 mL.

7.5 *Positive Displacement Pipette, and Tips to Suit*, 1 mL.

7.6 *Variable-Volume Dispenser*, 1 to 5 mL, fitted to a 1-L glass storage bottle.

8. Reagents and Materials

8.1 *Purity of Water*—Demineralised water as defined by Type I of Specification [D 1193](#).

8.2 *High Purity 0.3 M Nitric Acid Solution* (~x 50 dilution of the concentrated acid).

8.3 *Uranium Isotopic Reference Material (UIRMs)*—UIRMs are used to calibrate the instrument for multi-collection measurements. The Institute for Reference Materials and Measurements⁷ (IRMM) reference material IRMM-024 is used for enriched samples and the New Brunswick Laboratory⁸ Certified Reference Material CRM U005-A is used for samples of natural or depleted ^{235}U abundances. The UIRMs are prepared as uranyl nitrate solutions containing 0.4 µg/mL of uranium.

8.4 *Optional—Internal Reference Solution* containing ^{230}Th and ^{232}Th isotopes (or isotopes of another suitable element).

8.4.1 It has been found that the stability of the modern ICP-MC-MS can be such that it is not necessary to use an internal reference to monitor variations in mass bias. The data presented in this paper was obtained without the use of an internal reference. However, if the addition of an internal reference is deemed necessary then isotopes of thorium (230 and 232) can be used as a suitable internal reference material. The internal reference must contain at least one pair of isotopes in a fixed ratio. It is not necessary for this isotopic ratio to be accurately known as the same reference is added to both the calibration material and the subsequent samples. Minor fluctuations in instrument calibration (mass bias) are reflected in the measured ratio of the internal reference in the samples. Subsequent correction of the mass bias parameter using the measured ratio of the internal reference provides the necessary adjustment to the mass bias factor prior to result calculation.

8.4.2 The internal reference material should be prepared with a dilution appropriate to the sensitivity of the mass

⁶ The data presented in the paper was obtained using a ‘Nu Plasma’ mass spectrometer, manufactured by Nu Instruments (Nu Instruments Ltd, Unit 74 Clywedog Road South, Wrexham LL13 9XS, North Wales, UK). The Nu Plasma was supplied with the (optional) BIG80 vacuum pumping system to achieve optimum sensitivity.

⁷ Institute for Reference Materials and Measurement, Retieseweg, B-2440 Geel, Belgium.

⁸ New Brunswick Laboratory, D-350, 9800 South Cass Avenue, Argonne, Illinois 60439.

⁵ This correction can only be applied to samples which do not contain ^{239}Pu (or any other nuclides with mass 239).

TABLE 1

Collector	L6	L5	L4	IC1	L3	IC0	L2	L1	Ax	H1	H2	H3	H4	H5
Separation	2U	1U	1U	1U	1U	1U	1U	1U	1U	1U	1U	2U	2U	2U
Ion Beam	²³⁰ Th	²³² Th	–	²³⁴ U	²³⁵ U	²³⁶ U	–	²³⁸ U	²³⁸ UH	–	–	–	–	–

where:

- Ax = Axial Faraday collector,
 L and H = low and high mass Faraday collectors (with respect to the Axial collector),
 IC = ion counters, and
 U = unit mass dispersion for uranium isotopes.

spectrometer. If thorium is used as the internal reference then a thorium to uranium ratio of approximately 1:2 should be adequate.

NOTE 1—If an internal reference is added, then the uranic concentration of the samples should be adjusted so that the uranic concentration required for the mass spectrometer is achieved following the addition of the internal reference.

NOTE 2—The decay of ²³⁴U to ²³⁰Th may present a problem with the analysis of aged-uranic solutions. This should not present a problem with uranium hexafluoride samples that are taken in the gaseous phase, as gaseous UF₆ separates from any non-volatile thorium compounds.

9. Hazards

9.1 A number of the materials used in this procedure are radioactive, toxic, corrosive or any combination of the three. Adequate laboratory facilities and safe handling procedures must be used. A detailed discussion of all safety procedures is beyond the scope of this method. Site specific practices for the handling of radioactive materials and hazardous chemicals should be followed.

10. Sampling, Test Specimens, and Test Units ⁹

10.1 Samples Received as UF₆:

10.1.1 Transfer between 0.2 g and 0.25 g of UF₆ gas into a glass sample tube cooled by liquid nitrogen.¹⁰

10.1.2 Working in a fume cupboard, hydrolyse the UF₆ using demineralised water from a wash bottle. The operator should keep the sample tube pointed away at all times since some toxic HF gas is produced.

10.1.3 Pour the hydrolysed UF₆ into a 50 mL screw-cap polypropylene tube and dilute so that the final concentration of UF₆ is 5 mg/mL. For example, if the weight of UF₆ transferred is 0.2 g, dilute to 40 mL with demineralised water.

10.1.4 Using a positive displacement pipette, take a 0.01 mL aliquot of solution and transfer to a clean 50 mL screw-cap polypropylene tube. Dilute to a volume of 42 mL using a 0.3 M nitric acid solution. The resulting solution contains 1.2 µg/mL of UF₆ which is equivalent to 0.8 µg/mL of uranium.

10.1.5 Pour 2 mL of solution into a 10 mL polypropylene tube and double the volume to 4 mL using 0.3 M nitric acid solution, to reduce the uranic concentration to 0.4 µg/mL.

10.1.6 If required, add an aliquot of the thorium internal reference and mix the solution thoroughly (see 8.4).

⁹ The sample dilutions specified in this section can be varied according to instrument requirements.

¹⁰ Subsampling of UF₆ is detailed in ASTM Standard Test Method C 761.

10.1.7 Place the tube in the designated rack position in accordance with Section 13.

10.2 Samples Received as Aqueous Uranyl Nitrate Solutions of Known Uranic Concentration:

10.2.1 Dilute the sample with a 0.3 M nitric acid solution so that the uranium concentration is 0.8 µg/mL.

10.2.2 Proceed in accordance with 10.1.5-10.1.7.

11. Preparation of Apparatus

11.1 Many ICP-MC-MS designs require the Faraday collectors to be mechanically positioned to align with the ion beams. The instrument used for this work adopts a different approach, where a “zoom lens” which alters the dispersion of the instrument is used to focus the beams onto a fixed array of detectors. The zoom lens settings were adjusted under software control to achieve the configuration shown in Table 1.

11.2 To minimise measurement uncertainty, minor isotope (²³⁴U and ²³⁶U) abundances are measured with ion counters. The analyser magnet must be calibrated across the mass range 230 to 238, however, the instrument manufacturer recommends calibrating across the mass range 80 to 238 (achieved using the beam from the Argon dimer). The magnet should be re-calibrated if the calibration drifts by more than 0.2 atomic mass units.

12. Calibration and Standardization

12.1 Calibration of the mass spectrometer using a UIRM produces a mass bias factor. The mass bias factor for the UIRM in question is defined in Eq 2:

$$\text{Mass Bias Factor} = \left(\frac{\frac{^{235}\text{U}}{^{238}\text{U}} \text{ quoted}}{\frac{^{235}\text{U}}{^{238}\text{U}} \text{ measured}} \right)^{\frac{1}{\Delta m}} \quad (2)$$

where

Δm = ratio mass difference (that is, 3 in the case of the ²³⁵U/²³⁸U ratio).

12.2 The mass bias factor is exported to all subsequent sample measurements to correct for mass bias effects. Details of how the mass bias correction is applied can be found in Appendix X1. As stated in Section 8, IRMM-024 is used to calibrate for mass bias for samples enriched in ²³⁵U and NBL CRM 005-A is used to calibrate for mass bias for natural samples or samples depleted in ²³⁵U. Stock solutions of both these uranium reference materials (containing 0.4 µg/mL of uranium in a 0.3 M nitric acid solution) are held in the

laboratory. Mass bias calibration is an integral part of each sample run (that is, no separate calibration procedure is necessary).

13. Procedure

13.1 The instrument software is used to assign sample names, measurement procedures and auto-sampler rack positions to both UIRMs and samples. Two sample racks are used. The first rack contains the UIRM IRMM-024 and those samples that are enriched in ²³⁵U. The second rack contains the UIRM NBL CRM U005-A and those samples at natural level or depleted in ²³⁵U. The plasma is then struck and after a period of 60 minutes (to allow the system to thermally stabilise), a tuning solution of UIRM IRMM-024 is aspirated and the ion beams are optimised. The automatic measurement sequence is then initiated under computer control.

14. Calculation or Interpretation of Results

14.1 The measurement software produces three mass bias corrected ratios as follows:

$$r_4 = {}^{234}\text{U}/{}^{238}\text{U} \quad r_5 = {}^{235}\text{U}/{}^{238}\text{U} \quad r_6 = {}^{236}\text{U}/{}^{238}\text{U}$$

14.2 The ²³⁵U abundance is calculated from the following expression, where the figure of 1 is to account for the ²³⁸U/²³⁵U ratio:

$${}^{235}\text{U} = \frac{r_5}{1 + r_4 + r_5 + r_6} \times 100 \text{ \% atomic} \quad (3)$$

similarly

$${}^{234}\text{U} = \frac{r_4}{1 + r_4 + r_5 + r_6} \times 100 \text{ \% atomic} \quad (4)$$

and

$${}^{236}\text{U} = \frac{r_6}{1 + r_4 + r_5 + r_6} \times 100 \text{ \% atomic} \quad (5)$$

14.3 Finally the ²³⁸U % atomic is calculated by difference:

$${}^{238}\text{U} = 100 - ({}^{234}\text{U} + {}^{235}\text{U} + {}^{236}\text{U}) \text{ \% atomic} \quad (6)$$

14.4 The isotopic abundances in % atomic can be expressed as % mass using the following equations:

$${}^{235}\text{U} = \frac{b \times B}{(a \times A) + (b \times B) + (c \times C) + (d \times D)} \times 100 \text{ \% mass} \quad (7)$$

where:

a = ²³⁴U % atomic

b = ²³⁵U % atomic

c = ²³⁶U % atomic

d = ²³⁸U % atomic

A = atomic weight of ²³⁴U (234.0409)

B = atomic weight of ²³⁵U (235.0439)

C = atomic weight of ²³⁶U (236.0457)

D = atomic weight of ²³⁸U (238.0508)

similarly

$${}^{234}\text{U} = \frac{a \times A}{(a \times A) + (b \times B) + (c \times C) + (d \times D)} \times 100 \text{ \% mass} \quad (8)$$

and

$${}^{236}\text{U} = \frac{c \times C}{(a \times A) + (b \times B) + (c \times C) + (d \times D)} \times 100 \text{ \% mass} \quad (9)$$

15. Precision and Bias

15.1 *Precision*—The intermediate precision of the method for multi-collection measurements at various ²³⁵U abundances is presented in the following tables. Certified uranium abundances are quoted with 2 sigma confidence intervals. The units of precision are % Atomic (see Table 2).

15.2 *Bias*—In practice the observed agreement (at the 2 sigma level) between the certified and measured isotopic abundances for NBS050, NBS030, NBS020 and NBS005 (see Table 2) indicates that there is no significant bias.

15.3 *Limit of Detection*—The limit of detection was derived from measurements of ²³⁶U in samples derived from natural uranium (that is, samples with negligible ²³⁶U content). The results of 10 determinations of ²³⁶U both on natural uranium and an enriched (3.3 % ²³⁵U) material derived from natural are summarised as follows. Each determination is based on an individual sample measurement. The 10 determinations were distributed over several measurement runs (by more than one operator).

²³⁵ U Abundance (% Atomic)	²³⁵ U Limit of Detection 3 × Standard Deviation (% Atomic)
0.72	0.000001
3.32	0.000008

TABLE 2 Precision

Uranium Isotope	Certified Abundance (% Atomic)	Mean Measured Abundance (% Atomic)	Precision, (2s)	Relative Precision, %
10 Determinations of NBS050 ^A				
234	0.0279 ± 0.0001	0.0279	0.00018	0.65
235	5.010 ± 0.005	5.0097	0.0018	0.04
236	0.0480 ± 0.0002	0.0480	0.00015	0.31
10 Determinations of NBS030 ^A				
234	0.0190 ± 0.0001	0.01907	0.00008	0.42
235	3.046 ± 0.003	3.0460	0.0014	0.05
236	0.0204 ± 0.0001	0.02041	0.00014	0.69
10 Determinations of NBS020 ^A				
234	0.0125 ± 0.0001	0.01246	0.00004	0.32
235	2.038 ± 0.002	2.0374	0.0013	0.06
236	0.0165 ± 0.0001	0.01655	0.00009	0.54
10 Determinations of NBS005 ^A				
234	0.00218 ± 0.00004	0.00221	0.00001	0.45
235	0.4895 ± 0.0005	0.48911	0.00030	0.06
236	0.00466 ± 0.00005	0.00478	0.000050	1.05
Uranium Isotope	Accepted Abundance (% Atomic)	Mean Measured Abundance (% Atomic)	Precision (2s)	Relative Precision %
10 Determinations of UCL QC Natural ^B				
234	0.00530	0.00531	0.00001	0.23
235	0.7200	0.7199	0.00083	0.12
236	0.00008	0.00008	0.000003	4.3
10 Determinations of UCL QC Depleted ^B				
234	0.00182	0.00186	0.000016	0.9
235	0.3353	0.3355	0.0006	0.18
236	0.00763	0.00772	0.00004	0.5

^A NBS standards supplied by New Brunswick Laboratory in the form of U₃O₈.

^B UCL QC designates an "in-house," well characterised, bulk quality control material, derived from UF₆.

16. Keywords

16.1 ICP-MC-MS; ICP-MS; inductively coupled plasma mass spectrometer; internal reference; isotope ratio; isotopic abundance; mass bias; mass bias factor; mass bias parameter; memory; multi-collector; thorium; uranium

APPENDIX

(Nonmandatory Information)

X1. MASS BIAS CORRECTION AND RELATIVE COMBINED UNCERTAINTIES (WITH NO INTERNAL REFERENCE)

X1.1 Let q_5 be the quoted value of the $^{235}\text{U} / ^{238}\text{U}$ ratio for the uranium isotopic reference material (UIRM) and p_5 be the measured value of the $^{235}\text{U} / ^{238}\text{U}$ ratio for the UIRM. Let f = mass bias factor per amu.

X1.2 Mass bias (f) is defined as: $f^{\Delta M} = \frac{q_n}{p_n}$ and, therefore, in this case $f = \left(\frac{q_5}{p_5}\right)^{1/3}$ where ΔM is the isotopic mass difference for the quoted or measured ratio in question.

now

$$f = q_5^{1/3} p_5^{-1/3}$$

and

$$\delta f = \frac{\partial f}{\partial q_5} \delta q_5 + \frac{\partial f}{\partial p_5} \delta p_5$$

therefore

$$\frac{\partial f}{\partial q_5} = \frac{1}{3} q_5^{-2/3} p_5^{-1/3}$$

and

$$\frac{\partial f}{\partial p_5} = -\frac{1}{3} q_5^{1/3} p_5^{-4/3}$$

so

$$\delta f = \frac{1}{3} q_5^{-2/3} p_5^{-1/3} \delta q_5 - \frac{1}{3} q_5^{1/3} p_5^{-4/3} \delta p_5$$

and

$$\frac{\delta f}{f} = \frac{1}{3} \frac{\delta q_5}{q_5} - \frac{1}{3} \frac{\delta p_5}{p_5}$$

The most probable value of $\delta f/f$ is given by:

$$\frac{\delta f}{f} = \sqrt{\left(\frac{1}{3} \frac{\delta q_5}{q_5}\right)^2 + \left(\frac{1}{3} \frac{\delta p_5}{p_5}\right)^2} \quad (\text{X1.1})$$

In Eq X1.1, $\frac{\delta q_5}{q_5}$ represents the relative uncertainty quoted for the UIRM used in the calibration. The second term $\frac{\delta p_5}{p_5}$ represents the relative measurement uncertainty in the mass bias factor “ f ”. For replicate measurements of a given sample (or QC reference) where mass bias has been determined prior to each measurement of that sample, then the measurement uncertainty in the mass bias factor is reflected in the uncertainty of the isotope ratios measured for that sample. For such measurements Eq X1.1 simplifies to:

$$\frac{\delta f}{f} = \frac{1}{3} \frac{\delta q_5}{q_5}$$

X1.2.1 *Subsequent Samples:*

let

$$r_5 = ^{235}\text{U} / ^{238}\text{U} = m_5 f^3 \quad (\text{X1.2})$$

$$r_4 = ^{234}\text{U} / ^{238}\text{U} = m_4 f^4 \quad (\text{X1.3})$$

$$r_6 = ^{236}\text{U} / ^{238}\text{U} = m_6 f^2 \quad (\text{X1.4})$$

where m_n is a measured ratio, and r_n is m_n corrected for mass bias derived from the above calibration.

now

$$\delta r_5 = \frac{\partial r_5}{\partial f} \delta f + \frac{\partial r_5}{\partial m_5} \delta m_5$$

therefore

$$\delta r_5 = 3 f^2 m_5 \delta f + f^3 \delta m_5$$

and

$$\frac{\delta r_5}{r_5} = 3 \frac{\delta f}{f} + \frac{\delta m_5}{m_5}$$

Then the most probable value of $\frac{\delta r_5}{r_5}$ is given by:

$$\frac{\delta r_5}{r_5} = \sqrt{\left(3 \frac{\delta f}{f}\right)^2 + \left(\frac{\delta m_5}{m_5}\right)^2} \quad (\text{X1.5})$$

In Eq X1.5, $\frac{\delta f}{f}$ represents the relative uncertainty in the mass bias factor.

The second term, $\frac{\delta m_5}{m_5}$ represents the relative uncertainty in the measured $^{235}\text{U} / ^{238}\text{U}$ ratio.

Likewise, the most probable value of $\frac{\delta r_4}{r_4}$ is given by:

$$\frac{\delta r_4}{r_4} = \sqrt{\left(4 \frac{\delta f}{f}\right)^2 + \left(\frac{\delta m_4}{m_4}\right)^2} \quad (\text{X1.6})$$

In Eq X1.6, $\frac{\delta f}{f}$ represents the relative uncertainty in the mass bias factor.

The second term, $\frac{\delta m_4}{m_4}$ represents the relative uncertainty in the measured $^{234}\text{U} / ^{238}\text{U}$ ratio.

Likewise, the most probable value of $\delta r_6/r_6$ is given by:

$$\frac{\delta r_6}{r_6} = \sqrt{\left(2 \frac{\delta f}{f}\right)^2 + \left(\frac{\delta m_6}{m_6}\right)^2} \quad (\text{X1.7})$$

In Eq X1.7, δr_n represents the relative uncertainty in the mass bias factor.

The second term, $\delta m_6/m_6$ represents the relative uncertainty in the measured $^{236}\text{U}/^{238}\text{U}$ ratio.

X1.2.2 Isotopic Abundances:

The ratios r_4 , r_5 and r_6 are used to calculate the abundances of the uranium isotopes. For uranium isotope “n” (that is, ^{238}U and r_n), the abundance of ^{238}U is given by C_n , where:

$$\text{Abundance, } C_n = \frac{r_n}{1 + r_4 + r_5 + r_6} \text{ and let } D = 1 + r_4 + r_5 + r_6$$

also let

$$D = r_n + S$$

where

$$S = \sum r_x - r_n \text{ and } \sum r_x = 1 + r_4 + r_5 + r_6$$

then

$$C_n = \frac{r_n}{r_n + S}$$

now

$$\delta C_n = \frac{\partial C_n}{\partial r_n} \delta r_n + \frac{\partial C_n}{\partial S} \delta S$$

but

$$\frac{\partial C_n}{\partial r_n} = \frac{S}{D^2} \text{ and } \frac{\partial C_n}{\partial S} = -\frac{r_n}{D^2}$$

therefore

$$\delta C_n = \frac{1}{D^2} (S \delta r_n - r_n \delta S)$$

Because the ratios are all dependent on the value of ^{238}U , a worse case scenario is considered for assessing the uncertainty, δS , in S (that is, the dependent variables are fully correlated).

$$\delta S = \sum \delta r_x - \delta r_n$$

δr_n is an inherent component of $\sum \delta r_x$. Therefore, to compute δS , it is necessary to subtract the δr_n component from $\sum \delta r_x$.

Again, because r_n and S are dependent variables a worse case scenario is considered for assessing the uncertainty, δC_n , in C_n .

therefore

$$\delta C_n = \frac{1}{D^2} (S \delta r_n + r_n \delta S) \quad (\text{X1.8})$$

now

$$C_n = \frac{r_n}{D}$$

Dividing the LHS of Eq X1.8 by C_n and the RHS of Eq X1.8 by $\frac{r_n}{D}$ gives:

$$\begin{aligned} \frac{\delta C_n}{C_n} &= \frac{1}{r_n D} (S \delta r_n + r_n \delta S) \\ \frac{\delta C_n}{C_n} &= \frac{1}{D} \left(S \frac{\delta r_n}{r_n} + \delta S \right) \end{aligned} \quad (\text{X1.9})$$

Substituting for $\frac{\delta C_n}{C_n}$, D , S , $\frac{\delta r_n}{r_n}$ and δS in Eq X1.9 for each uranium isotope gives:

$$\frac{\delta C_5}{C_5} = \frac{1}{D} \left((1 + r_4 + r_6) \frac{\delta r_5}{r_5} + \delta r_4 + \delta r_6 \right) \quad (\text{X1.10})$$

$$\frac{\delta C_4}{C_4} = \frac{1}{D} \left((1 + r_5 + r_6) \frac{\delta r_4}{r_4} + \delta r_5 + \delta r_6 \right) \quad (\text{X1.11})$$

$$\frac{\delta C_6}{C_6} = \frac{1}{D} \left((1 + r_4 + r_5) \frac{\delta r_6}{r_6} + \delta r_4 + \delta r_5 \right) \quad (\text{X1.12})$$

$$\frac{\delta C_8}{C_8} = \frac{1}{D} (\delta r_4 + \delta r_5 + \delta r_6) \quad (\text{X1.13})$$

Eq X1.10-X1.13 are used to calculate the relative uncertainty in each of the uranium isotopic abundances. However, for Eq X1.11 and Eq X1.12 the values of $\frac{\delta C_4}{C_4}$ and $\frac{\delta C_6}{C_6}$ both need to be increased by an additional 0.01 to account for the uncertainty quoted for the minor isotopes in the reference material used to calibrate the gains of the associated ion counters.

Using the preceding equations and the data from Section 15, estimates of the combined uncertainties of measurement for multi-collection measurements of ^{234}U , ^{235}U , and ^{236}U at various abundances were computed. The estimates are presented in the tables below. All uncertainties are percent relative expanded uncertainties at the 95 % confidence level.

^{235}U Abundance % Atomic	Overall Relative Expanded Uncertainty (%)	Data Used for Uncertainty Assessment
0.20–0.50	0.30	NBS005, QCD
0.5–1.00	0.10	QCN
1.00–6.0	0.08	NBS 020, 030, 050

^{234}U or ^{236}U Abundance % Atomic	Overall Relative Expanded Uncertainty
< 0.0050	0.0001 absolute
≥ 0.0050	1 %

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