



Standard Test Method for Isotopic Abundance Analysis of Uranium Hexafluoride 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, multi-collector, mass spectrometry (ICP-MC-MS). This test method is also described in ASTM STP 1344.

1.2 *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:

C 761 Test Methods for Chemical, Mass Spectrometric, Spectrochemical, Nuclear, and Radiochemical Analysis of Uranium Hexafluoride²

C 787 Specification for Uranium Hexafluoride for Enrichment²

C 996 Specification for Uranium Hexafluoride Enriched to less than 5 % ^{235}U ²

D 1193 Specification for Reagent Water³

2.2 Other Document:

STP 1344 Applications of Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) to Radionuclide Determinations⁴

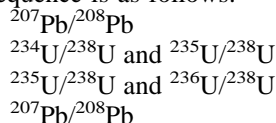
3. Summary of Test Method

3.1 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. Subsequently, an internal reference of either thorium or lead isotopes⁵ is added to each diluted sample.

3.2 The samples are contained in polythene 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.

3.3 Uranium isotopic reference materials (UIRMs) are used to calibrate the instrument. Each UIRM is prepared in aqueous solution (acidified with nitric acid) and 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 measurements⁶ to correct the measured uranium isotopic ratios.

3.4 Measurement of isotopic ratios in the calibration solution and the subsequent samples is initiated by customized software. The measurement sequence is determined by the internal reference used. Using the lead internal reference, the sequence is as follows:



Using the mean of the two lead ratios and the mass bias parameter imported from the calibration, the current mass bias factor is computed. The mass bias factor is then used to correct the measured uranium isotopic ratios. These corrected ratios are used to calculate the abundances of ^{234}U , ^{235}U , and ^{236}U .

3.5 Using the thorium internal reference, the sequence is as follows:



Using the $^{230}\text{Th}/{}^{232}\text{Th}$ ratios (that are acquired simultaneously to the $^{234}\text{U}/{}^{238}\text{U}$ and $^{235}\text{U}/{}^{238}\text{U}$ ratios) and the mass bias parameter imported from the calibration, the mass bias factor is computed. The mass bias factor is then used to correct the the measured $^{234}\text{U}/{}^{238}\text{U}$ and $^{235}\text{U}/{}^{238}\text{U}$ ratios in “real time.” The $^{236}\text{U}/{}^{238}\text{U}$ is retrospectively corrected for mass bias. The abundances are expressed as % atomic. A printout of results and mass spectrometer parameters is obtained for each sample. Details of the mass bias correction are presented in Appendix X1.

⁶ The uranium isotopic reporting limits and reporting errors are listed in section 16 and Appendix X1 respectively.

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 Annual Book of ASTM Standards, Vol 12.01.

³ Annual Book of ASTM Standards, Vol 11.01.

⁴ Available from ASTM Headquarters.

⁵ Thorium is preferred as the internal reference because thorium data can be acquired simultaneously to uranium data.

4. Significance and Use

4.1 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.

5. Interferences

5.1 *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.

5.2 *Adjacent Isotopic Peaks*—The abundance sensitivity of the ICP-MC-MS at mass 237 is specified to be less than 2 parts per million of the ^{238}U ion beam. For all enrichment plant samples, the ^{235}U isotopic abundance is currently no more than 6 % mass, consequently, interference effects with the ^{234}U and ^{236}U ion beams are negligible.

5.3 *Isobaric Molecular Interferences*—Any $^{235}\text{UH}^+$ ions formed in the plasma produce an interference with the ^{236}U ion beam. The magnitude of the $^{235}\text{UH}^+$ ion has been assessed by measuring the mass 236 peak of a natural uranium reference material containing no ^{236}U . The ratio of ^{235}UH to ^{235}U was recorded and subsequently used to correct ^{236}U measurements.

5.4 Memory Effects:

5.4.1 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 2 % nitric acid solution and measuring the background ^{238}U ion beam. If the ^{238}U background ion beam exceeds $1\text{E-}14$ amps, then the sample introduction system is stripped and cleaned.

5.4.2 If thorium is used as the internal reference, then it is possible to correct for uranium background (^{235}U and ^{238}U only) by measuring a (blank) solution of 2 % nitric acid spiked with the thorium isotopes. In practice, it has been found unnecessary to apply a background correction to UIRMs or samples enriched in ^{235}U .

6. Apparatus

6.1 Mass Spectrometer:

6.1.1 The mass spectrometer has an inductively coupled plasma (ICP) source and a double-focussing electrostatic/magnetic sector analyser equipped with seven Faraday detectors and one Daly detector.⁷

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

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

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

6.4 *Fixed-Volume Pipette and Tips to Suit*, 0.01 mL.

6.5 *Fixed-Volume Pipette and Tips to Suit*, 1 mL.

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

7. Reagents

7.1 *Purity of Water*—Demineralised water as defined by Type I of Specification D 1193.

7.2 *ICP Standard Pb Solution* (1000 ppm).

7.3 *Nitric Acid Solution*, analytical grade, various concentrations. Necessary dilutions can be inferred from the stated acid strength (for example, 2 % nitric acid solution requires a $\times 50$ dilution of the concentrated acid).

7.4 *Reference Solution* containing 140 ppb of ^{230}Th ⁸ and 7 ppm of ^{232}Th .

7.5 *Uranium Isotopic Reference Materials*.

8. Internal References

8.1 *Requirements*—As described in Section 3, either lead or thorium can be used as an internal reference to be added to the UIRMs and uranium samples. 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.

8.2 *Lead*—If lead is used as the internal reference, the $^{207}\text{Pb}/^{208}\text{Pb}$ ratio is monitored. The lead is prepared by using a 2 % nitric acid solution to dilute a 1000- $\mu\text{g/mL}$ lead stock standard to 7 $\mu\text{g/mL}$. The dilute lead solution is stored in a 1-L bottle fitted with an adjustable dispenser set to 1 mL. A 1-mL aliquot of the lead solution is dispensed into 6 mL of sample solution containing 1.2 $\mu\text{g/mL}$ of uranium. The resultant concentration of the both the uranium and lead internal reference is 1 $\mu\text{g/mL}$.

8.3 *Thorium*—If thorium is used as the internal reference, the $^{230}\text{Th}/^{232}\text{Th}$ ratio is monitored. The thorium solution is prepared by adding ^{230}Th to a calculated quantity of ^{232}Th from a 1000- $\mu\text{g/mL}$ stock standard which is then diluted with 2 % nitric acid. The quantity of ^{230}Th added is such that the final diluted reference should contain 140 ng/mL of ^{230}Th and 7 $\mu\text{g/mL}$ of ^{232}Th . This solution is stored in a 1-L bottle fitted with an adjustable dispenser set to 1 mL. A 1-mL aliquot of thorium solution is dispensed into 6 mL of sample solution containing 1.2 $\mu\text{g/mL}$ of uranium. The resultant concentration of ^{230}Th is 20 ng/mL and the resultant concentration of ^{232}Th is 1 $\mu\text{g/mL}$.

NOTE 1—The ^{230}Th is radioactive (α -emitter) and consequently, the quantity of ^{230}Th is minimised to comply with local disposal safety regulations.

9. Uranium Isotopic Reference Materials (UIRMs)

9.1 UIRMs are used to calibrate the instrument for multi-collection measurements. The Institute for Reference Materials and Measurements⁹ (IRMM) reference material IRMM-024 is

⁸ ^{230}Th is supplied by AEA Technology, Harwell, Didcot, Oxfordshire, UK.

⁹ Institute for Reference Materials and Measurement, Retieseweg, B-2440 Geel, Belgium

⁷ The VG Elemental, Plasma54 (P54) is such a mass spectrometer.

used for enriched samples and the New Brunswick Laboratory¹⁰ Certified Reference Material CRM U005-A is used for samples of natural or depleted ²³⁵U abundances. The UIRMs are prepared as uranyl nitrate solutions containing 1.2 µg/mL of uranium and the same quantity of internal reference as described in Section 8.

10. Instrument Setup

10.1 Choice of internal reference determines the position of the adjustable Faraday collector buckets. Buckets are positioned under software control. If lead is chosen as the internal reference, then the distances between the collectors are tabulated as follows:

Collector ^A Separation ^A	L2	L1	Ax	H1	H2	H3	H4
	1Pb	1U	1U	1U	1U	1U	1U

^AKey to Symbols:

Ax = axial collector.

H = collector on high-mass side of the axial collector.

L = collector on low-mass side of the axial collector.

1U = unit mass dispersion for uranium isotopes.

2U = twice unit mass dispersion for uranium isotopes.

1Pb = unit mass dispersion for lead isotopes.

If thorium is chosen as the internal reference, then the distances between the collectors are again tabulated as follows:

Collector ^A Separation ^A	L2	L1	Ax	H1	H2	H3	H4
	2U	2U	1U	1U	1U	1U	1U

^AKey to Symbols:

Ax = axial collector.

H = collector on high-mass side of the axial collector.

L = collector on low-mass side of the axial collector.

1U = unit mass dispersion for uranium isotopes.

2U = twice unit mass dispersion for uranium isotopes.

1Pb = unit mass dispersion for lead isotopes.

Either a Daly or Faraday detector can be selected as the axial collector. To optimize measurement uncertainty, all minor isotope (²³⁴U and ²³⁶U) measurements are on the Axial (Daly) collector. The analyser magnet must be calibrated across the mass range from 207 to 238. The magnet must be re-calibrated if the calibration drifts by more than the 0.2 atomic mass units (at uranium).

11. Sample and Blank Preparation

11.1 Samples Received as UF₆:

11.1.1 Transfer between 0.2 and 0.25 g of UF₆ gas into a glass sample tube cooled by liquid nitrogen.¹¹

11.1.2 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 toxic HF gas is produced.

11.1.3 Pour the hydrolyzed 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 demineralized water.

11.1.4 Using a fixed-volume 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 28 mL using a 2 % nitric acid solution. The resulting solution contains 1.8 µg/mL of UF₆ which is equivalent to 1.2 µg/mL of uranium.

¹⁰ New Brunswick Laboratory, D-350, 9800 South Cass Avenue, Argonne, Illinois 60439.

¹¹ Subsampling of UF₆ is detailed in ASTM Standard Test Method C 761.

11.1.5 Pour 6 mL of solution into a 10-mL polypropylene tube.

11.1.6 Add 1 mL of the 7-µg/mL internal reference and thoroughly mix the solution.

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

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

11.2.1 Dilute the sample with a 2 % nitric acid solution so that the uranium concentration is 1.2 µg/mL

11.2.2 Proceed in accordance with 11.1.5-11.1.7.

11.3 Blank Solution (used only if thorium is the internal reference):

11.3.1 Pour 6 mL of 2 % nitric acid solution into a 10-mL polythene tube.

11.3.2 Add 1 mL of the 7-µg/mL thorium internal reference and thoroughly mix the solution.

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

12. Calibration

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 1.

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

where

Δm = ratio mass difference (that is, 3).

12.2 The mass bias factor is applied to the measured isotope ratio of the internal reference to produce a mass bias parameter. This parameter 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 9, 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 1.2 µg/mL of uranium in a 2 % 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. Operational Procedure

13.1 The instrument software is used to assign sample names, measurement procedures, and auto-sampler rack positions to UIRMs and samples. Two sample racks are used. Rack A contains the UIRM IRMM-024, a tuning solution, and those samples that are enriched in ²³⁵U. Rack B contains the blank (if thorium is the internal reference), a 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 30 min (to allow the system to thermally stabilize), the tuning solution is aspirated and the ion beams are optimized. Blank measurement (if required), mass bias calibration, and sample measurement are then initiated under computer control.

14. Calculation

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

$$r_4 = \frac{234}{238} \quad r_5 = \frac{235}{238} \quad r_6 = \frac{236}{238}$$

The ^{235}U abundance is calculated from the following expression:

$$^{235}\text{U} = \frac{r_5}{1 + r_4 + r_5 + r_6} \times 100 \% \text{ atomic} \quad (2)$$

similarly

$$^{234}\text{U} = \frac{r_4}{1 + r_4 + r_5 + r_6} \times 100 \% \text{ atomic} \quad (3)$$

and

$$^{236}\text{U} = \frac{r_6}{1 + r_4 + r_5 + r_6} \times 100 \% \text{ atomic} \quad (4)$$

Finally the ^{238}U % atomic is calculated by difference:

$$^{238}\text{U} = 100 - (^{234}\text{U} + ^{235}\text{U} + ^{236}\text{U}) \% \text{ atomic} \quad (5)$$

Before reporting, the isotopic abundancies in % atomic are converted to % mass.

$$^{235}\text{U} = \frac{b \times B}{(a \times A) + (b \times B) + (c \times C) + (d \times D)} \times 100 \% \text{ mass} \quad (6)$$

where:

$a = ^{234}\text{U} \% \text{ atomic}$

$b = ^{235}\text{U} \% \text{ atomic}$

$c = ^{236}\text{U} \% \text{ atomic}$

$d = ^{238}\text{U} \% \text{ atomic}$

$A = \text{atomic weight of } ^{234}\text{U} (234.0409)$

$B = \text{atomic weight of } ^{235}\text{U} (235.0439)$

$C = \text{atomic weight of } ^{236}\text{U} (236.0457)$

$D = \text{atomic weight of } ^{238}\text{U} (238.0508)$

similarly

$$^{234}\text{U} = \frac{a \times A}{(a \times A) + (b \times B) + (c \times C) + (d \times D)} \times 100 \% \text{ mass} \quad (7)$$

and

$$^{236}\text{U} = \frac{c \times C}{(a \times A) + (b \times B) + (c \times C) + (d \times D)} \times 100 \% \text{ mass} \quad (8)$$

15. Precision and Bias

15.1 *Precision*—The precision results are quoted from measurements using lead as the internal reference. However, during the instrument commissioning trials a thorium internal reference was used and comparable precisions were obtained. Four analysts from the same laboratory were involved in the measurement program. The intermediate precision of the method for multi-collection measurements at various ^{235}U abundances is presented in the following tables. Certified

uranium isotopic abundances are quoted with 2 sigma confidence intervals. The units of precision are % Atomic (see Table 1).

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 (tabulated in 15.1) indicates that there is no significant bias.

16. Limit of Detection

16.1 The limit of detection was derived from measurements of ^{236}U in samples derived from natural uranium (that is, samples with negligible ^{236}U content). The results of 22 determinations of ^{236}U both on natural uranium and an enriched (3.3 % ^{235}U) material derived from natural are summarized as follows. Each determination is based on an individual sample measurement. The 22 determinations were distributed over several measurement runs (by more than one operator) during the course of a month. Consequently, the standard deviation quoted is representative of intermediate precision.

^{235}U Abundance, % Atomic	^{236}U Limit of Detection $3 \times$ Standard Deviation, % Atomic
0.72	0.00002
3.32	0.00004

17. Keywords

17.1 Internal reference; Isotope ratio; lead; mass bias; mass bias factor; mass bias parameter; memory; multi-collector, inductively coupled plasma source mass spectrometer (MC-ICP-MS); thorium; uranium

TABLE 1 Precision

22 Determinations of NBS050 ^A Using Multi-Collection Faraday/Daly				
Uranium Isotope	Certified Abundance, % Atomic	Mean Measured Abundance, % Atomic	Precision, (2s)	Relative Precision, %
234	0.0279 ± 0.0001	0.02789	0.00008	0.27
235	5.010 ± 0.005	5.0085	0.0013	0.026
236	0.0480 ± 0.0002	0.04813	0.00013	0.27
150 Determinations of NBS030 ^A Over Six Months Using Multi-Collection Faraday/Daly				
234	0.0190 ± 0.0001	0.01905	0.00007	0.37
235	3.046 ± 0.003	3.0462	0.0010	0.034
236	0.0204 ± 0.0001	0.02047	0.00008	0.39
22 Determinations of NBS020 ^A Using Multi-Collection Faraday/Daly				
234	0.0125 ± 0.0001	0.01244	0.00006	0.46
235	2.038 ± 0.002	2.0375	0.00065	0.032
236	0.0165 ± 0.0001	0.01650	0.00006	0.38
130 Determinations of NBS005 ^A Over Six Months Using Multi-Collection Faraday/Daly				
234	0.00218 ± 0.00004	0.00219	0.00001	0.49
235	0.4895 ± 0.0005	0.4894	0.0004	0.073
236	0.00466 ± 0.00005	0.00474	0.00003	0.56
22 Determinations of UCL QC ^B Natural Using Multi-Collection Faraday/Daly				
234	0.0053	0.00525	0.00004	0.77
235	0.7199	0.72004	0.00033	0.046
236	0.00003	0.00005	0.00001	26.1
22 Determinations of UCL QC ^B Depleted Using Multi-Collection Faraday/Daly				
234	0.0018	0.00180	0.00003	1.4
235	0.3350	0.33510	0.00022	0.066
236	0.0076	0.00756	0.00005	0.61

^ADesignated NBS as supplied.

^BUCL QC designates an "in-house," well-characterised, bulk quality control material.

APPENDIX

(Nonmandatory Information)

X1. DERIVATION OF RELATIVE COMBINED UNCERTAINTIES

X1.1 For the following equations, a = mass bias parameter/amu and b = mass bias factor/amu.

X1.2 For the uranium isotopic reference material (UIRM) used in the calibration q_n = quoted value of the ratio $^{23}\text{n}/^{238}$ and p_n = measured value of the ratio $^{23}\text{n}/^{238}$. The unit a is defined as follows:

$$a = \left(\frac{q_5}{p_5}\right)^{1/3} \left(\frac{^{207}\text{Pb}}{^{208}\text{Pb}}\right) \text{ (measured in UIRM)} \quad (\text{X1.1})$$

or

$$a = \left(\frac{q_5}{p_5}\right)^{2/3} \left(\frac{^{230}\text{Th}}{^{232}\text{Th}}\right) \text{ (measured in UIRM)} \quad (\text{X1.2})$$

Then for the subsequent samples, b is defined as follows:

$$b = a \left(\frac{^{207}\text{Pb}}{^{208}\text{Pb}}\right)^{-1} \text{ (measured in sample)} \quad (\text{X1.3})$$

or

$$b = \sqrt{a \left(\frac{^{230}\text{Th}}{^{232}\text{Th}}\right)^{-1}} \text{ (measured in sample)} \quad (\text{X1.4})$$

Let $x = b^3$, $y = b^4$, $z = b^2$, and $m_n = \frac{^{23}\text{n}}{^{238}}$ (measured ratio)

Then

$$r_5 = m_5 b^3 = m_5 x \quad (\text{X1.5})$$

$$r_4 = m_4 b^4 = m_4 y \quad (\text{X1.6})$$

$$r_6 = m_6 b^2 = m_6 z \quad (\text{X1.7})$$

The mass bias factor, b , is raised to the power of the ratio mass difference, and r_n is a ratio that has been corrected for mass bias derived from the calibration.

Let $C_5 = ^{235}\text{U}$ abundance.

$$\text{Hence } C_5 = \frac{r_5}{1 + r_4 + r_5 + r_6}$$

$$\text{and } \delta C_5 = \frac{\partial C_5}{\partial r_5} \delta r_5 + \frac{\partial C_5}{\partial r_4} \delta r_4 + \frac{\partial C_5}{\partial r_6} \delta r_6$$

But

$$\frac{\partial C_5}{\partial r_5} = \frac{1 + r_4 + r_6}{D^2} \quad (\text{X1.8})$$

where

$$D = 1 + r_4 + r_5 + r_6$$

and

$$\frac{\partial C_5}{\partial r_4} = \frac{\partial C_5}{\partial r_6} = \frac{-r_5}{D^2} \quad (\text{X1.9})$$

Hence

$$\frac{\partial C_5}{C_5} = \frac{1}{D} \sqrt{\left(\frac{\delta r_5}{r_5}\right)^2 + \delta r_4^2 + \delta r_6^2} \quad (\text{X1.10})$$

From Eq X1.5, the corrected ratio, r_5 , is comprised of two independent components, the measured ratio and the mass bias correction factor as follows:

$$r_5 = m_5 x \quad (\text{X1.11})$$

Each component has an associated uncertainty and therefore the most probable uncertainty, δr_5 , is given as follows:

$$\frac{\delta r_5}{r_5} = \sqrt{\left(\frac{\delta m_5}{m_5}\right)^2 + \left(\frac{\delta x}{x}\right)^2} \quad (\text{X1.12})$$

where

$$\frac{\delta x}{x} = \sqrt{\left(\frac{\delta q_5}{q_5}\right)^2 + \left(\frac{\delta p_5}{p_5}\right)^2} \quad (\text{X1.13})$$

Similarly from Eq X1.6 and X1.7 are as follows:

$$r_4 = m_4 y \quad (\text{X1.14})$$

and

$$r_6 = m_6 z \quad (\text{X1.15})$$

Therefore

$$\delta r_4 = r_4 \sqrt{\left(\frac{\delta m_4}{m_4}\right)^2 + \left(\frac{\delta y}{y}\right)^2} \quad (\text{X1.16})$$

and

$$\delta r_6 = r_6 \sqrt{\left(\frac{\delta m_6}{m_6}\right)^2 + \left(\frac{\delta z}{z}\right)^2} \quad (\text{X1.17})$$

Now

$$y = b^4 = \left(\frac{q_5}{p_5}\right)^{4/3} \quad (\text{X1.18})$$

and

$$\delta y = \frac{\partial y}{\partial q_5} \delta q_5 + \frac{\partial y}{\partial p_5} \delta p_5 \quad (\text{X1.19})$$

$$\therefore \delta y = \frac{4}{3} \left(\frac{q_5}{p_5}\right)^{1/3} \frac{1}{p_5} \delta q_5 - \frac{4}{3} \left(\frac{q_5}{p_5}\right)^{1/3} \frac{q_5}{p_5^2} \delta p_5 \quad (\text{X1.20})$$

$$\therefore \frac{\delta y}{y} = \frac{4}{3} \left(\frac{q_5}{p_5}\right)^{1/3} \left(\frac{q_5}{p_5}\right)^{-4/3} \frac{\delta q_5}{p_5} - \frac{4}{3} \left(\frac{q_5}{p_5}\right)^{1/3} \left(\frac{q_5}{p_5}\right)^{-4/3} \frac{q_5 \delta p_5}{p_5^2} \quad (\text{X1.21})$$

$$\therefore \frac{\delta y}{y} = \frac{4}{3} \left(\frac{\delta q_5}{q_5} - \frac{\delta p_5}{p_5}\right) \quad (\text{X1.22})$$

As q_5 and p_5 are independent entities, then the most probable relative uncertainty, $\frac{\delta y}{y}$, is as follows:

$$\frac{\delta y}{y} = \frac{4}{3} \sqrt{\left(\frac{\delta q_5}{q_5}\right)^2 + \left(\frac{\delta p_5}{p_5}\right)^2} \quad (\text{X1.23})$$

Similarly

$$z = b^2 = \left(\frac{q_5}{p_5}\right)^{2/3} \quad (\text{X1.24})$$

Then using the same analysis technique as for $\frac{\delta y}{y}$,

Substituting Eq X1.12, Eq X1.16, and Eq X1.17 into Eq X1.10 gives

$$\frac{\delta z}{z} = \frac{2}{3} \sqrt{\left(\frac{\delta q_5}{q_5}\right)^2 + \left(\frac{\delta p_5}{p_5}\right)^2} \quad (\text{X1.25})$$

$$\frac{\delta C_5}{C_5} = \frac{1}{D} \sqrt{(1 + r_4 + r_6)^2 \left(\left(\frac{\delta m_5}{m_5} \right)^2 + \left(\frac{\delta x}{x} \right)^2 \right) + r_4^2 \left(\left(\frac{\delta m_4}{m_4} \right)^2 + \left(\frac{\delta y}{y} \right)^2 \right) + r_6^2 \left(\left(\frac{\delta m_6}{m_6} \right)^2 + \left(\frac{\delta z}{z} \right)^2 \right)} \quad (\text{X1.26})$$

Similarly

$$\frac{\delta C_4}{C_4} = \frac{1}{D} \sqrt{(1 + r_5 + r_6)^2 \left(\left(\frac{\delta m_4}{m_4} \right)^2 + \left(\frac{\delta y}{y} \right)^2 \right) + r_5^2 \left(\left(\frac{\delta m_5}{m_5} \right)^2 + \left(\frac{\delta x}{x} \right)^2 \right) + r_6^2 \left(\left(\frac{\delta m_6}{m_6} \right)^2 + \left(\frac{\delta z}{z} \right)^2 \right)} \quad (\text{X1.27})$$

and

$$\frac{\delta C_6}{C_6} = \frac{1}{D} \sqrt{(1 + r_4 + r_5)^2 \left(\left(\frac{\delta m_6}{m_6} \right)^2 + \left(\frac{\delta z}{z} \right)^2 \right) + r_4^2 \left(\left(\frac{\delta m_4}{m_4} \right)^2 + \left(\frac{\delta y}{y} \right)^2 \right) + r_5^2 \left(\left(\frac{\delta m_5}{m_5} \right)^2 + \left(\frac{\delta x}{x} \right)^2 \right)} \quad (\text{X1.28})$$

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 Table 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
0.3–0.5	0.099	NBS005 and QCD
0.7	0.082	QCN
2.0	0.068	NBS020
3.0	0.068	NBS030
5.0	0.064	NBS050

For purposes of reporting, the expanded uncertainties in table are rounded up to 0.1 %:

^{234}U or ^{236}U Abundance	Overall Relative Expanded Uncertainty
<50 ppm	1ppm absolute
≥50 ppm	1 %

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