

Standard Test Method for Isotopic Analysis of Uranium Hexafluoride by Single–Standard Gas Source Multiple Collector Mass Spectrometer Method¹

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1. Scope

1.1 This test method is applicable to the isotopic analysis of uranium hexafluoride (UF₆) with 235 U concentrations less than or equal to 5 % and ^{234}U , ^{236}U concentrations of 0.0002 to 0.1 %.

1.2 This test method may be applicable to the analysis of the entire range of ²³⁵U isotopic compositions providing that adequate Certified Reference Materials (CRMs or traceable standards) are available.

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 health practices and determine the applicability of regulatory limitations prior to use.

2. Referenced Documents

2.1 ASTM Standards:²

- C787 Specification for Uranium Hexafluoride for Enrichment
- C996 Specification for Uranium Hexafluoride Enriched to Less Than 5 % ²³⁵U
- 2.2 Other Document:
- USEC-651 Uranium Hexafluoride: A Manual of Good Handling Practices³

3. Summary of Test Method

3.1 The unknown sample and a CRM or traceable standard whose isotopic composition is close to that of the sample are introduced in sequence into the mass spectrometer, and ²³⁴ U,

²³⁵U, ²³⁶U, and ²³⁸U ions are focused through corresponding collector slits to the four separate collectors. Measurements are made that are proportional to the ratios of ²³⁴U, ²³⁵U, or ²³⁶U to ²³⁸U. With the known composition of the CRM or traceable standard, these ratios of molar ratios permit calculation of the ²³⁴U, ²³⁵ U, and ²³⁶U contents. Memory corrections are applied based on the periodic measurement of two CRMs or traceable standards.

4. Significance and Use

4.1 Uranium hexafluoride is a basic material used to produce nuclear reactor fuel. To be suitable for this purpose, the material must meet criteria for isotopic composition. This test method is designed to determine whether the material meets the requirements described in Specifications C787 and C996.

5. Apparatus

5.1 A gas source multiple collector mass spectrometer with the following attributes:

5.1.1 The resolving power of the mass spectrometer is not less than 500. The resolving power (R) is calculated from the registered mass spectrum of both the $^{235}\text{UF}_5^+$ and $^{238}\text{UF}_5^+$ isotopes as follows:

$$R = \frac{a \cdot M}{b \cdot \Delta M} \tag{1}$$

where

- = distance between centers of the ${}^{235}\text{UF}_5^+$ and ${}^{238}\text{UF}_5^+$ a peaks,
- $b = \text{peak width of the} \, {}^{238}\text{UF}_5^+ \text{ isotope (10 \% valley),} \\ M = 333 \text{mass(u)} \, {}^{238}\text{UF}_5^+, \text{ and} \\ \Delta M = 3 = 333 330, \, 330 \text{mass(u)} \, {}^{235}\text{UF}_5^+$

5.1.2 The abundance sensitivity of the mass spectrometer is specified as less than 1×10^{-5} as contribution of mass 333 $(^{238}UF_5^+)$ to mass 331 $(^{236}UF_5^+)$.

5.1.3 The four collectors have collector slits adjusted for ions of masses 329, 330, 331, and 333. Ion currents are amplified by four amplifiers, having noise level less than 0.5 mV.

5.1.4 The ion beams are kept within the slits by an automatic beam positioner circuit.

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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.

Available from United States Enrichment Corporation, 6903 Rockledge Drive, Bethesda, MD 20817.

5.1.5 The pumping system of the mass spectrometer analyser tube shall maintain a pressure less than 5×10^{-8} torr with a sample flowing into the ion source.

5.1.6 The memory correction factor of the mass spectrometer as defined in 9.1 shall be consistent with the required accuracy and precision, and shall not exceed 1.005.

5.1.7 The sample inlet system shall be equipped with a manifold, including adjustable leak, and valves for introducing the sample and CRM or traceable standard in sequence and for evacuating corresponding lines. The pumping system of the inlet system must maintain a pressure less than 2×10^{-2} torr by evacuating.

6. Hazards

6.1 Since UF_6 is radioactive, toxic, and highly reactive, especially with reducing substances and moisture (see USEC-651), appropriate facilities and practices for analysis shall be provided.

7. Calibration and Standardization

7.1 Uranium Hexafluoride Isotopic

7.1.1 Two CRMs or traceable standards are used to determine the memory correction factor. The ²³⁵U concentration ratio of upper CRM or traceable standard (C_{235}^{SH}) to ²³⁵U of lower CRM or traceable standard (C_{235}^{SL}) shall not be more than three ($C_{235}^{SH}/C_{235}^{SL} \le 3$).

7.1.2 For memory correction factor determination for ²³⁴U and ²³⁶U isotopes, two CRMs or traceable standards are used which differ in ²³⁴ U (²³⁶U) concentration. In concentration range 0.0002 to 0.01, $C_{234 (236)}^{SH}/C_{234 (236)}^{SL} \leq 8$; in concentration range 0.01 to 0.1, $C_{234 (236)}^{SH} \leq 6$.

7.1.3 The CRMs or traceable standards used for measurements may differ in ²³⁵U concentration from a sample. (C_{235}^x) on condition that $C_{235}^{SH}/C_{235}^x \le 1.5$ and $C_{235}^x/C_{235}^{SL} \le 1.5$. For ²³⁴U (²³⁶U) isotopes, the following range limitations shall be used: 0.0002 to 0.01 $C_{234(236)}^{SH}/C_{234(236)}^x \le 4, C_{234(236)}^x/C_{234(236)}^{SL} \le 3, C_{234(236)}^x/C_{234(236)}^{SL} \le 3$.

8. Procedure

A typical sequence for the analytical determination is X, S, X, S, where X and S mean the introduction of the sample and the CRM or traceable standard, respectively. Each introduction is followed by ion source evacuation before the next introduction. During each introduction, a simultaneous measurement of the four uranium isotopes occurs.

The intensities of the ${}^{238}\text{UF}_5^+$ ion beam for both sample and CRM or traceable standard introduction shall not differ more than 3 %. Adjustment is performed by pressure equalization of the sample and standard in the inlet system.

The number of introductions per analytical sequence is dependent on the precision required.

8.1 Attach sample containers containing the appropriate sample, X, and standard, S, to the inlet system, and prepare both materials for introduction into the ion source as follows:

8.1.1 Operate the appropriate valves to remove air entrapped in the connectors and to check that there are no leaks in inlet system. 8.1.2 Freeze the UF_6 by immersing the sample container (the unknown sample) into a mixture of water and ice.

8.1.3 Open the valve on the container to permit evacuation of volatile impurities, and then close the valve.

8.1.4 Remove the coolant from around the container and allow the UF_6 to return to room temperature.

8.1.5 Repeat 8.1.2-8.1.4 for the CRM or traceable standard.

8.2 Operation of the Mass Spectrometer

8.2.1 Operate appropriate valves to admit the CRM or traceable standard into the ion source.

8.2.2 Adjust the accelerating voltage or magnet current to focus the ion beams $^{234}\rm{UF}_5^+$, $^{235}\rm{UF}_5^+$, 236 UF $_5^+$, and $^{238}\rm{UF}_5^+$ to their corresponding collectors. Adjust the mass spectrometer parameters to obtain the maximum $^{238}\rm{UF}_5^+$ ion current and maximum resolution.

8.2.3 Regulate the adjustable leak to obtain a $^{238}\text{UF}_5^+$ ion current of about 10^{-9} A.

8.2.4 Measure the ion current ratio of 234 U, 235 U, and 236 U to 238 U.

8.2.5 Terminate the flow of the CRM or traceable standard and evacuate the ion source.

8.2.6 Repeat 8.2.1, 8.2.4, and 8.2.5 for the sample.

9. Calculation

9.1 The memory correction factor M is calculated by the formula:

$$M_{i} = \frac{\frac{E_{i}^{S1} - E_{i}^{S2}}{E_{i}^{S2}}}{\frac{r_{i}^{S1} - r_{i}^{S2}}{r_{i}^{S2}}}$$
(2)

where:

 M_i = memory correction factor,

- $E_i^{s_1}, E_i^{s_2}$ = molar ratios of isotope of interest to ²³⁸U is calculated from certified data of the two CRMs or traceable standards taken for memory determination,
- r_i^{S1} = measured ion current ratio of isotope of interest to ²³⁸U for CRM or traceable standard 1, and

 r_i^{S2} = measured ion current ratio of isotope of interest to 238 U for CRM or traceable standard 2.

9.2 The ratio of molar ratios for the CRMs or traceable standards is calculated as follows:

$$R_i^s = \frac{r_i^{s_1}}{r_i^{s_2}}$$
(3)

9.3 The ratio of molar ratios for the sample is calculated as follows:

$$R_i^X = \frac{r_i^X}{r_i^S} \tag{4}$$

where:

- r_i^X = measured ion current ratio of isotope of interest to ²³⁸U for the sample, and
- r_i^S = measured ion current ratio of isotope of interest to ²³⁸U for the working CRM or traceable standard.

9.4 The corrected molar ratio E_i^x is calculated for the sample using the memory correction factor, M_i , and certified data of

			Mass-spectrometer A			Mass-spectrometer B		
	CRMs	CRM 7533-99, 207	-05 C	CRM 7523-99, 210-06	CRM 7533-99	, 207-05	CRM 7520-99, 3-79	
	used for determination of	(Working CRM)				(Working CRM)		
	the memory correction factor	$C_{235}^{S} = 4.9512 \pm 0.0024$	$C_{235}^S = 1.2$	2047±0.0005	$C_{235}^S = 4.9512 \pm 0.0024$	$C_{235}^{S} =$	0.5040±0.0003	
Reg. No. RM, Lot number, Certified values,		$C_{234}^{S} = 0.04612 \pm 0.00015$	$C_{234}^{S} = 0.0$	00747±0.00002	$C_{234}^{S} = 0.04612 \pm 0.00015$	$C_{234}^{S} =$	0.0035±0.0001	
		$C_{236}^{S} = 0.01521 \pm 0.00010$	$C_{236}^{S} = 0.0$	00231±0.00002	$C_{236}^{s} = 0.01521 \pm 0.00010$	$C_{236}^{S} =$	0.0060±0.0001	
	Isotope Mass Fraction, %							
	CRMs		CRM 7530-99, 186-01			CRM 7530-99, 186-01		
	used as a sample	C_{235}^{χ} C_{234}^{χ}	1	C_{236}^{χ}	C_{235}^{χ}	C_{234}^{χ}	C_{236}^{χ}	
	Reg. No. RM, Lot number,	4.0032 ± 0.0019	0.0360 ± 0.0002	0.0133 ± 0.0002	4.0032 ± 0.0019	0.0360 ± 0.0002	0.0133 ± 0.0002	
	Certified values,							
	Isotope Mass Fraction, %							
Mean (\overline{C}_{i}^{x})		4.0024	0.03590	0.01324	4.0035	0.03595	0.01328	
(-,)	Quantity of determinations	9	9	9	10	10	10	
	under repeatability (n)	-						
	Relative standard	0.006	0.12	0.43	0.006	0.09	0.33	
	uncertainty. %							
Bias $(\overline{C}^{X} - C^{X})$	x)	0.0008	0.00010	0.00006	0.0003	0.00005	0.00005	
	CBMs	CBM 7533-99 207	'-05 C	BM 7523-99 210-06	CRM 7533-99	207-05	CBM 7520-99 3-79	
	used for determination of	(Working CBM)	(Morking CPM)			(Morking CDM)		
	the memory correction factor	$C^{s} = 4.9512 \pm 0.0024$	$C^{S} = 1.2$	$C^{S} = 1.2047 \pm 0.0005$		$C^{S} = 4.9512 \pm 0.0024$ $C^{S} = 0.5040 \pm 0.0002$		
	Reg No BM Lot number	$C_{235}^{S} = 0.04612 \pm 0.00015$	$C_{235}^{S} = 0.0$	2047 ± 0.00000	$C_{235}^{S} = 0.04612 \pm 0.0024$	$C_{235}^{S} = C_{235}^{S}$	0.0025±0.0001	
	Cortified values	$C_{234}^{S} = 0.04612 \pm 0.00015$	$C_{234}^{\circ} = 0.0$	00747 ± 0.00002	$C_{234}^{\circ} = 0.04612 \pm 0.00013$	$C_{234} = C_{234}^{S} = C_{234}^{S}$	0.0035 ± 0.0001	
	Lectone Mass Fraction %	$C_{236}^{-} = 0.01521 \pm 0.00010$	$U_{236}^{-} - 0.0$	J0231±0.00002	C ₂₃₆ 0.01521±0.00010	U ₂₃₆ -	0.0000±0.0001	
			ODM 7507 00, 010 00			CPM 7527 00 210 08		
			CRIVI 7527-99, 219-08		CX	CNIVI 7527-99, 219-	CX CX	
	used as a sample		0.00545 . 0.0001					
	Reg. No. RM, Lot number,	2.8250 ± 0.0011	0.02545 ± 0.0001	0.00070 ± 0.00003	2.8250 ± 0.0011	0.02545 ± 0.0001	0.00070 ± 0.00003	
	Certified values,							
(<u>a</u> v)	isotope mass Fraction, %	0.0040	0.00540	0.00070	0.0040	0.00544	0.00067	
Mean (C_i^{\star})		2.8248	0.02542	0.00072	2.8243	0.02544	0.00067	
	Quantity of determinations	10	10	10	10	10	10	
	under repeatability (n)							
	Relative standard	0.005	0.13	5.2	0.009	0.14	7.2	
	uncertainty, %							
Bias ($\overline{C}_{i}^{X} - C_{i}^{Z}$	x)	0.0002	0.00003	0.00002	0.0007	0.00001	0.00003	
	CRMs	CRM 7520-99, 3-	79 C	CRM 7523-99, 210-06	CRM 7520-9	9, 3-79	CRM 7523-99, 210-06	
	used for determination of			(Working CRM)	(Working CRM)			
	the memory correction factor	$C_{235}^{S} = 0.5040 \pm 0.0003$	$C_{235}^{S} = 1.2$	$C_{235}^{S} = 1.2047 \pm 0.0005$		$C_{235}^{S} = 0.5040 \pm 0.0003$ $C_{235}^{S} = 1.2047 \pm 0.0005$		
	Reg. No. RM, Lot number,	$C_{234}^{S} = 0.0035 \pm 0.0001$	$C_{234}^{S} = 0.0$	00747±0.00002	$C_{234}^{S} = 0.0035 \pm 0.0001$	$C_{234}^{S} =$	0.00747±0.00002	
	Certified values,	$C_{236}^{S} = 0.0060 \pm 0.0001$	$C_{236}^{S} = 0.00231 \pm 0.00002$		$C_{236}^{S} = 0.0060 \pm 0.0001$	$C_{236}^{S} =$	0.00231±0.00002	
	Isotope Mass Fraction, %	200				200		
	CRM		CRM 7521-99, 184-00		CRM 7521-99, 184-00			
	used as a sample	C_{235}^{χ} C_{234}^{χ}	1	C_{236}^{χ}	C_{235}^{χ}	C_{234}^{χ}	C_{236}^{χ}	
	Reg. No. RM, Lot number,	0.71049 ± 0.00029	0.00531 ± 0.00007	< 0.00002	0.71049 ± 0.00029	0.00531 ± 0.00007	< 0.00002	
	Certified values,							
	Isotope Mass Fraction, %							
Mean $\left(\overline{C}_{i}^{x}\right)$	• •	0.710496	0.00533		0.710463	0.005312		
	Quantity of determinations	10	10		10	10		
	under reneatability (n)		10		10	10		
	Polativo standard	0.03	0.7		0.02	0.8		
		0.03	0.7		0.02	0.8		
(=-	uncertainty, %	0.000000	0.00000		0 00007	0.000000		
Bias $\left(C_{i}^{x} - C \right)$	<u>^)</u>	0.00006	0.00002		0.000027	0.000002		

TABLE 1 Uranium Isotopic CRMs Results

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the CRM or traceable standard:

$$E_i^X = \left[1 + M_i \cdot \left(R_i^X - 1\right)\right] \cdot E_i^S \tag{5}$$

where:

 E_i^s = ratio of certified molar concentrations of isotope of interest to ²³⁸U for the working CRM or traceable standard.

9.5 Calculate the molar concentration of isotope of interest for the sample:

$$C_i^{\mathrm{X}} = \frac{E_i^{\mathrm{X}} \cdot 100}{1 + \sum E_j^{\mathrm{X}}} \tag{6}$$

where:

 E_j^X = corrected molar ratios for ²³⁴U, ²³⁵U, and ²³⁶U (see 9.4).

9.6 Calculate the weight percent for 234 U, 235 U, and 236 U as follows:

$$C_i = \frac{C_i^{\mathsf{X}} \cdot A_i}{\sum C_i^{\mathsf{X}} \cdot A_i} \tag{7}$$

where:

 C_i^X = molar concentration of the isotope *j*,

 A_i = atomic mass of the isotope *i* of interest, and

 A_i = atomic masses of the isotope *j*.

10. Precision and Bias

10.1 Isotopic uranium CRMs have been analyzed in 2009. The measurements were conducted in one laboratory on two mass spectrometers by various operators.

10.2 *Precision*—For each CRM, the average measured value, \overline{C}_i^x , is given together with the relative standard uncertainty, obtained for *n* experiments under repeatability. Results are listed in Table 1.

10.3 *Bias*—In accordance with Table 1, the method shows no statistically significant bias.

11. Keywords

11.1 gas source multiple collector mass spectrometer; uranium hexafluoride isotopic CRM or traceable standard; uranium isotopes

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