Designation: C 1428 – 99

Standard Test Method for Isotopic Analysis of Uranium Hexafluoride by Single–Standard Gas Source Multiple Collector Mass Spectrometer Method[1]

This standard is issued under the fixed designation C 1428; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision. A number in parentheses indicates the year of last reapproval. A superscript epsilon (ϵ) indicates an editorial change since the last revision or reapproval.

1. Scope

1.1 This test method is applicable to the isotopic analysis of uranium hexafluoride (UF_6) with ^{235}U concentrations less than or equal to 5 % and ^{234}U, ^{236}U concentrations of 0.001 to 0.1 %.

1.2 This test method may be applicable to the analysis of the entire range of ^{235}U isotopic compositions providing that adequate 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:*

C 787 Specification for Uranium Hexafluoride for Enrichment[2]

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

2.2 *Other Document:*

USEC-651 Uranium Hexafluoride: A Manual of Good Handling Practices[3]

3. Summary of Test Method

3.1 The unknown sample and a standard whose isotopic composition is close to that of the sample are introduced in sequence into the mass spectrometer, and ^{234}U, ^{235}U, ^{236}U, and ^{238}U ions are focused through corresponding collector slits to the four separate collectors. Measurements are made that are proportional to the ratios of ^{234}U, ^{235}U, or ^{236}U to ^{238}U. With the known composition of the standard, these ratios of molar ratios permit calculation of the ^{234}U, ^{235}U, and ^{236}U contents. Memory corrections are applied based on the periodic measurement of two 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 C 787 and C 996.

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 $^{235}UF_5^+$ and $^{238}UF_5^+$ isotopes as follows:

$$R = \frac{a \cdot M}{b \cdot \Delta M} \quad (1)$$

where

a = distance between centers of the $^{235}UF_5^+$ and $^{238}UF_5^+$ peaks,
b = peak width of the $^{238}UF_5^+$ isotope (10 % valley),
M = 333 − mass(u) $^{238}UF_5^+$, and
ΔM = 3 = 333 − 330, 330 − mass(u) $^{235}UF_5^+$

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.

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 1.01.

5.1.7 The sample inlet system shall be equipped with a manifold, including adjustable leak, and valves for introducing the sample and standard in sequence and for evacuating corresponding lines. The pumping system of the inlet system

[1] This test method is under the jurisdiction of ASTM Committee C–26 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]

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 standards are used to determine the memory correction factor. The relative ^{235}U concentration difference of these standards shall not be more than 20 %.

7.1.2 For memory correction factor determination for ^{234}U and ^{236}U isotopes in concentration range 0.01 to 0.1 %, two standards are used which differ in ^{234}U (^{236}U) concentration. The relative difference shall not exceed two times; in concentration range 0.001 to 0.01 %, the relative difference shall not exceed four times.

7.1.3 The standards used for measurements may differ in ^{235}U concentration from a sample, but the relative difference shall not be more than 10 %. The relative difference in ^{234}U (^{236}U) concentration shall not exceed two times for the concentration range 0.001 to 0.01 %, and shall not be more than 50 % for the concentration range 0.01 to 0.1 %.

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 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 $^{238}U_5^+$ ion beam for both sample and 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 standard.

8.2 *Operation of the Mass Spectrometer*

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

8.2.2 Adjust the accelerating voltage or magnet current to focus the ion beams $^{234}UF_5^+$, $^{235}UF_5^+$, $^{236}UF_5^+$, and $^{238}UF_5^+$ to their corresponding collectors. Adjust the mass spectrometer parameters to obtain the maximum $^{238}UF_5^+$ ion current and maximum resolution.

8.2.3 Regulate the adjustable leak to obtain a $^{238}UF_5^+$ 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 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:

$$M_i = \frac{\dfrac{E_i^{S1} - E_i^{S2}}{E_i^{S2}}}{\dfrac{r_i^{S1} - r_i^{S2}}{r_i^{S2}}} \tag{2}$$

where:

M_i = memory correction factor,
E_i^{S1}, E_i^{S2} = molar ratios of isotope of interest to ^{238}U is calculated from certified data of the two standards taken for memory determination,
r_i^{S1} = measured ion current ratio of isotope of interest to ^{238}U for Standard 1, and
r_i^{S2} = measured ion current ratio of isotope of interest to ^{238}U for Standard 2.

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

$$R_i^S = \frac{r_i^{S1}}{r_i^{S2}} \tag{3}$$

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 ^{238}U for the sample, and
r_i^S = measured ion current ratio of isotope of interest to ^{238}U for the working 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 the standard:

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

where:

E_i^S = ratio of certified molar concentrations of isotope of interest to ^{238}U for the working standard.

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

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

where:

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

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

$$C_i = \frac{C_i^X \cdot A_i}{\sum C_j^X \cdot A_j} \quad (7)$$

where:

C_j^X = molar concentration of the isotope *j*,
A_i = atomic mass of the isotope *i* of interest, and
A_j = atomic masses of the isotope *j*.

10. Precision and Bias

10.1 Isotopic uranium standards have been analysed from 1995 to 1996. The measurements were conducted in one laboratory on two mass spectrometers by various operators.

10.2 *Precision*—For each standard, the average measured value, X, is given together with the standard deviation, S_r, obtained for n experiments. Results are listed in Table 1.

The standards used for measurements differed in isotopic composition from the analyzed standards. The relative difference in ^{235}U concentration was equal close to 10 %, and one in^{234}U and ^{236}U concentrations was equal close to 50 %.

TABLE 1 Uranium Isotopic Standards Results

SRM	Certified Values ± absolute error	Measured Values X	S_r	n
^{235}U (atom %)				
95.148 − 90	0.71978 ± 0.00005	0.71983	0.00028	25
95.162 − 90	2.4377 ± 0.0012	2.4373	0.0005	20
95.167 − 90	3.1434 ± 0.0015	3.1428	0.0003	20
95.173 − 90	4.252 ± 0.002	4.2511	0.0004	20
^{234}U (atom %)				
95.148 − 90	0.00533 ± 0.00005	0.00532	0.000014	20
95.173 − 90	0.0326 ± 0.0002	0.0327	0.00007	20
^{236}U (atom %)				
95.173 − 90	0.0036 ± 0.0001	0.0036	0.00018	20
95.170 − 90	0.0199 ± 0.0003	0.0202	0.00020	20

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 standards; uranium isotopes