



Designation: C1429 – 21

# Standard Test Method for Isotopic Analysis of Uranium Hexafluoride by Double-Standard Multi-Collector Gas Mass Spectrometer<sup>1</sup>

This standard is issued under the fixed designation C1429; 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 ( $\epsilon$ ) indicates an editorial change since the last revision or reapproval.

## 1. Scope\*

1.1 This test method covers a quantitative test method applicable to determining the mass percent of uranium isotopes in uranium hexafluoride ( $\text{UF}_6$ ) samples. This method as described is for concentrations of  $^{235}\text{U}$  between 0.1 and 10 mass %, and  $^{234}\text{U}$  and  $^{236}\text{U}$  between 0.0001 and 0.1 mass %.

1.2 This test method is for laboratory analysis by a gas mass spectrometer with a multi-collector.

1.3 This standard complements Test Methods C761, the double-standard method for gas mass spectrometers using a single collector, by providing a method for spectrometers using a multi-collector.

1.4 *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, and environmental practices and determine the applicability of regulatory limitations prior to use.*

1.5 *This international standard was developed in accordance with internationally recognized principles on standardization established in the Decision on Principles for the Development of International Standards, Guides and Recommendations issued by the World Trade Organization Technical Barriers to Trade (TBT) Committee.*

## 2. Referenced Documents

### 2.1 ASTM Standards:<sup>2</sup>

C761 Test Methods for Chemical, Mass Spectrometric, Spectrochemical, Nuclear, and Radiochemical Analysis of Uranium Hexafluoride

C787 Specification for Uranium Hexafluoride for Enrichment

C859 Terminology Relating to Nuclear Materials

C996 Specification for Uranium Hexafluoride Enriched to Less Than 5 %  $^{235}\text{U}$

C1215 Guide for Preparing and Interpreting Precision and Bias Statements in Test Method Standards Used in the Nuclear Industry

### 2.2 Other Documents:

USEC 651 Good Handling Practices for Uranium Hexafluoride<sup>3</sup>

## 3. Terminology

### 3.1 Definitions:

3.1.1 Except as otherwise defined herein, definitions of terms are as given in Terminology C859.

### 3.2 Definitions of Terms Specific to This Standard:

3.2.1 *A standard, n*—the low-value standard of a standard pair that brackets the sample.

3.2.2 *B standard, n*—the high-value standard of a standard pair that brackets the sample.

3.2.3 *determination, n*—a single isotopic value, calculated from a sequence of ratios; the most basic isotopic value calculated.

3.2.4 *Lagrange's interpolation formula, n*—a mathematical equation designed to estimate values between two or more known values.

3.2.5 *run, n*—a completed, six-entry symmetrical sequence consisting of A standard, sample, B standard, B standard, sample, and A standard from which a determination can be calculated for one or more isotopes.

3.2.6 *standard spread, n*—the difference between the high and low standards; sometimes called standard range.

3.2.7 *test result, n*—a reported value; the mean of two or more determinations.

## 4. Summary of Test Method

4.1 Uranium hexafluoride gas is introduced into an ionization source. The resulting ions are accelerated down the flight tube into the magnetic field. The magnetic field separates the

<sup>1</sup> 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.

Current edition approved Oct. 1, 2021. Published November 2021. Originally approved in 1999. Last previous edition approved in 2014 as C1429 – 99 (2014). DOI: 10.1520/C1429-21.

<sup>2</sup> 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.

<sup>3</sup> Available from Centrus Energy Corp., 6901 Rockledge Drive, Suite 800 Bethesda, Maryland 20817, https://www.centrusenergy.com.

\*A Summary of Changes section appears at the end of this standard

ions into ion beams in accordance with the  $m/e$  ratio. Four collectors are stationed so the  $^{234}\text{UF}_5^+$ ,  $^{235}\text{UF}_5^+$ ,  $^{236}\text{UF}_5^+$ , and  $^{238}\text{UF}_5^+$  ion beams strike individual collectors.

4.2 Two standards are chosen whose values bracket the desired isotope of the sample. The sample and two standards are introduced in a six-entry, symmetrical sequence. Then, measurements are taken that give the mole ratio of the desired isotope to  $^{238}\text{U}$ .

4.3 Through Lagrange's interpolation formula, these measurements are used to calculate the mass percent of the desired isotope. If standards are available that bracket all isotopes, then the  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{236}\text{U}$  mass percents are calculated from the same six-entry run.

4.4 The results of two six-entry, symmetrical-sequence runs are averaged to find test results for each isotope. The  $^{238}\text{U}$  mass percent is obtained by subtraction.

## 5. Significance and Use

5.1 Uranium hexafluoride used to produce nuclear-reactor fuel must meet certain criteria for its isotopic composition. This test method may be used to help determine if sample materials meet the criteria described in Specifications **C787** and **C996**.

## 6. Apparatus

6.1 Mass spectrometer with the following features and capabilities:

6.1.1 An ion source with an accelerating voltage of approximately 8 kV,

6.1.2 A resolving power of greater than or equal to 500,

6.1.3 A minimum of three points of attachment for standards or samples,

6.1.4 An ion collection system consisting of four collector cups stationed to collect  $^{234}\text{UF}_5^+$ ,  $^{235}\text{UF}_5^+$ ,  $^{236}\text{UF}_5^+$ , and  $^{238}\text{UF}_5^+$  ions,

6.1.5 An ion-current amplifier for each collector cup,

6.1.6 A voltage-to-frequency (V-to-F) converter for each amplifier,

6.1.7 A counter for each V-to-F converter, and

6.1.8 Computer control over opening and closing valves, the timing, and the integration of analytical sequences.

## 7. Hazards

7.1 Uranium hexafluoride ( $\text{UF}_6$ ) is radioactive, toxic, and highly reactive, especially with reducing substances and moisture. Appropriate laboratory facilities, materials of construction, and techniques shall be utilized when handling  $\text{UF}_6$  (see, for example, USEC-651).

7.2 Follow all safety procedures for handling  $\text{UF}_6$  provided by the facility. Familiarization and compliance with the Safety Data Sheet is essential.

7.3 When released to atmosphere, gaseous  $\text{UF}_6$  reacts with moisture to produce hexafluoride (HF) gas and toxic  $\text{UO}_2\text{F}_2$  particulates. Use sufficient ventilation or respiratory protection

to avoid breathing fumes. Use appropriate personal protective equipment such as gloves, eye, and face protection.

## 8. Procedure

### 8.1 Select standards:

8.1.1 Choose high and low standards that bracket the sample isotope(s) being evaluated. If the mass percent of  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{236}\text{U}$  are all desired, then the two standards must bracket each of the three isotopes to permit calculation of all isotopes for every run.

8.1.2 If standards that bracket all isotopes are unavailable, analyze the isotope(s) bracketed by the originally selected standards, then select other standards to run the remaining isotope(s).

### 8.2 Prepare Sample and Standards:

8.2.1 Attach sample and standard containers to the spectrometer.

8.2.2 Open and close the appropriate valves to evacuate the air from the inlet system.

8.2.3 Open the sample and standard containers individually and vent the gas phase to the cold trap. This is to remove impurities that may bias the results or interfere with the ionization. If necessary, freeze the  $\text{UF}_6$  with ice water or a mixture of crushed dry ice and isopropyl alcohol to permit longer venting without losing large amounts of  $\text{UF}_6$ .

8.2.4 Permit exhaust system pressure to recover.

8.2.5 Check to see if impurities have been sufficiently removed by introducing  $\text{UF}_6$  into the ion source and observing pressure, or exhausting through the cold trap and observing pressure on the other side, or any other suitable means.

8.2.6 If necessary, repeat 8.2.3 – 8.2.5 until samples are clean.

### 8.3 Prepare Instrument:

8.3.1 Adjust instrument parameters to focus ion beams in proper collectors and maximize the  $^{238}\text{UF}_5^+$  current reading.

8.3.2 Enter standard values and other information if needed for calculations performed by computer.

8.3.3 Program the spectrometer to run two of the following six-entry, symmetrical sequences: low standard, sample, high standard, high standard, sample, low standard.

### 8.4 Run the Analysis:

8.4.1 Obtain measurements from all four collectors during each entry.

## 9. Calculation

9.1 Perform the following operations for each of the  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{236}\text{U}$  isotopes:

9.1.1 For each entry, obtain a ratio by dividing the  $\text{UF}_5^+$  ion count of the desired isotope by the  $^{238}\text{UF}_5^+$  ion count.

9.1.2 Find the mean of the two low standard ratios and designate this A.

9.1.3 Find the mean of the two sample ratios and designate this X.

9.1.4 Find the mean of the two high standard ratios and designate this B.

NOTE 1—In a six-entry symmetrical run sequence,

$$(r_1 + r_6)/2 = A \quad (1)$$

$$(r_2 + r_5)/2 = X \quad (2)$$

$$(r_3 + r_4)/2 = B \quad (3)$$

where:

$r_n$  = the ratio from the  $n^{\text{th}}$  entry.

9.1.5 Find the mass percent ratio of the low value standard ( $A$  standard) by dividing the mass percent of the desired isotope by the mass percent  $^{238}\text{U}$ .

$$E_A = \text{mass \% } ^{234}\text{U}/\text{mass \% } ^{238}\text{U} \quad (4)$$

$$H_A = \text{mass \% } ^{235}\text{U}/\text{mass \% } ^{238}\text{U} \quad (5)$$

$$Y_A = \text{mass \% } ^{236}\text{U}/\text{mass \% } ^{238}\text{U} \quad (6)$$

9.1.6 Find the equivalent mass percent ratio for the high value standard ( $B$  standard.) Label it either  $E_B$ ,  $H_B$ , or  $Y_B$ .

9.1.7 Find the difference ( $D$ ) between the mass percent ratios of the  $A$  and  $B$  standards.

NOTE 2— $E_B - E_A = D_E$ ,  $H_B - H_A = D_H$ , and  $Y_B - Y_A = D_Y$

9.1.8 Find the mass percent ratio (desired isotope/ $^{238}\text{U}$ ) of the sample by calculating  $E_X$ ,  $H_X$ , or  $Y_X$  as follows:

$$E_X = ((X - A)/(B - A)) \cdot D_E + E_A \quad (7)$$

$$H_X = ((X - A)/(B - A)) \cdot D_H + H_A \quad (8)$$

$$Y_X = ((X - A)/(B - A)) \cdot D_Y + Y_A \quad (9)$$

NOTE 3—Calculations in 9.1.8 are based on an algebraic manipulation of Lagrange's interpolation formula using two known values of a function.

9.2 Calculate the mass percent as follows:

$$e = 100\% \cdot E_X / (1 + E_X + H_X + Y_X) \quad (10)$$

$$x = 100\% \cdot H_X / (1 + E_X + H_X + Y_X) \quad (11)$$

$$y = 100\% \cdot Y_X / (1 + E_X + H_X + Y_X) \quad (12)$$

where:

$$e = \text{mass \% } ^{234}\text{U},$$

$$x = \text{mass \% } ^{235}\text{U}, \text{ and}$$

$$y = \text{mass \% } ^{236}\text{U}.$$

9.3 Average a minimum of two determinations for each test result.

9.4 Find the mass percent  $^{238}\text{U}$  by subtracting the mass percent of the other isotopes from 100 %.

## 10. Precision and Bias

10.1 Seven standards traceable to National Institute of Standards and Technology (NIST) were selected as reference values. Two pairs of standards, also traceable to NIST, were chosen to evaluate the  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{236}\text{U}$  mass percent values of the seven reference standards. These standard pairs were obtained by selecting a low standard and two high standards to create one standard spread that was narrower for all three of the desired isotopes and one standard spread that was wider for all three of the desired isotopes. The  $^{234}\text{U}$  and  $^{236}\text{U}$  reference values assigned to all of these standards were determined by thermal mass spectrometer (see Table 1) with the exception of the mass percent  $^{236}\text{U}$  value assigned to G015-1. This value was obtained on a single-collector gas-

TABLE 1 Reference Materials Used in the Test and Their Assigned Values

Identity	Function	$^{234}\text{U}$ mass %	$^{235}\text{U}$ mass %	$^{236}\text{U}$ mass %
G005	low std	0.0026	0.48347	0.0027
G006	reference	0.0040	0.61074	0.0060
G010-1	reference	0.0052	0.99151	0.0077
G012	reference	0.0096	1.2250	0.0152 <sup>A</sup>
G015-1	reference	0.0060	1.5139	0.0152 <sup>A</sup>
G020-1	reference	0.0156	2.0123	0.0039
G025	reference	0.0204 <sup>A</sup>	2.4719	0.0018 <sup>B</sup>
G030-1	reference	0.0224 <sup>A</sup>	3.0066	0.0040
G030	high std	0.0186	3.0072	0.0108
G050-1	high std	0.0264	4.9457	0.0369

<sup>A</sup> These values are outside the bracket of the narrow standard spread, and only the data obtained from the wider standard pair are used to determine the precision and bias.

<sup>B</sup> This value is not within the bracket of either standard pair, and the data obtained for this value were not used to determine the precision and bias. The certainty in the assigned values, expressed as  $3 \times \%$  RSD is  $\pm 2.2921\%$  for  $^{234}\text{U}$ ,  $\pm 0.0646\%$  for  $^{235}\text{U}$ , and  $\pm 2.7263\%$  for  $^{236}\text{U}$ .

TABLE 2 Precision and Bias Test Results in Mass Percent Units

Isotope	%Bias <sup>A</sup>	$S_r$ <sup>B</sup>	%RSD <sub>r</sub> <sup>C</sup>	$S_R$ <sup>D</sup>	%RSD <sub>R</sub> <sup>E</sup>
$^{235}\text{U}$	-0.003	0.00010	0.006	0.00014	0.010
$^{234}\text{U}$	-0.810	0.000032	0.411	0.000085	0.722
$^{236}\text{U}$	3.711	0.000026	0.344	0.000120	1.664

<sup>A</sup> %Bias = (bias/reference value)  $\times$  100 %

<sup>B</sup>  $S_r$  = repeatability standard deviation

<sup>C</sup> %RSD<sub>r</sub> = %RSD for repeatability

<sup>D</sup>  $S_R$  = reproducibility standard deviation

<sup>E</sup> %RSD<sub>R</sub> = %RSD for reproducibility

N = 1260 for  $^{235}\text{U}$ , 1080 for  $^{234}\text{U}$ , 900 for  $^{236}\text{U}$

phase mass spectrometer. The  $^{235}\text{U}$  reference values were assigned through a combination of thermal and gas-phase analysis.

10.2 The test deviated from single-day design because of the need for a large number of test results for statistical purposes. However, each set of data for a specific combination of reference value, standard pair, and mass spectrometer was obtained within a 24-h period. Each reference standard was run by double-standard method 60 consecutive times for each of two standard pair on each of three mass spectrometers. Every run yielded three determinations, one for each isotope. Two consecutive determinations were then averaged for a test result. This produced a test result for each isotope after every two consecutive runs. The completion of each reference standard analysis yielded 30 test results per combination of spectrometer and standard pair for each of the  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{236}\text{U}$  isotopes. The combined test data for all isotopes of all reference standards yielded 7560 test determinations and 3780 test results. These data were collected over a period of several months.

10.3 The one-analyst design was impractical because of the need to accumulate such a large amount of data in as short a time as possible. After a reference standard was attached to a spectrometer, all the data for that combination of reference standard and standard pair were obtained before the reference standard was detached from the spectrometer. The contribution to variation due to different analysts should have been minimal.