**Designation: C 1429 – 99**

# Standard Test Method for Isotopic Analysis of Uranium Hexafluoride by Double— Standard Multi—Collector Gas Mass Spectrometer[1]

This standard is issued under the fixed designation C 1429; 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 is a quantitative test method applicable to determining the mass percent of uranium isotopes in uranium hexafluoride ($UF_6$) samples. This method as described is for concentrations of $^{235}U$ between 0.1 and 10 mass percent, and$^{234}U$ and $^{236}U$ between 0.0001 and 0.1 mass percent.

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

1.3 This standard complements C 761, sections 35 through 40, 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 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 Hexafluoride[2]

C 787 Specification for Uranium Hexafluoride for Enrichment[2]

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

C 1215 Guide for Preparing and Interpreting Precision and Bias Statements in Test Method Standards Used in the Nuclear Industry[2]

## 3. Terminology

3.1 *Definitions of Terms Specific to This Standard:*

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

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

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

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

3.1.5 *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.

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

3.1.7 *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 ions into ion beams in accordance with the *m/e* ratio. Four collectors are stationed so the $^{234}UF_5{}^+$, $^{235}UF_5{}^+$, $^{236}UF_5{}^+$, and$^{238}UF_5{}^+$ 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 which give the mole ratio of the desired isotope to $^{238}U$.

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 $^{234}U$, $^{235}U$, and $^{236}U$ 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 $^{238}U$ 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

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[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.

materials meet the criteria described in Specifications C 787 and C 996.

## 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 $^{234}UF_5^+$, $^{235}UF_5^+$, $^{236}UF_5^+$, and $^{238}UF_5^+$ 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. Procedure

7.1 *Select standards*

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

7.1.2 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).

7.2 *Prepare sample and standards*

7.2.1 Attach sample and standard containers to the spectrometer.

7.2.2 Open and close the appropriate valves to evaluate the air from the inlet system.

7.2.3 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 $UF_6$ with ice water or a mixture of crushed dry ice and isopropyl alcohol to permit longer venting without losing large amounts of $UF_6$.

7.2.4 Permit exhaust system pressure to recover.

7.2.5 Check to see if impurities have been sufficiently removed by introducing $UF_6$ 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.

7.2.6 If necessary, repeat 7.2.3-7.2.5 until samples are clean.

7.3 *Prepare instrument*

7.3.1 Adjust instrument parameters to focus ion beams in proper collectors and maximize the $^{238}UF_5^+$ current reading.

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

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

7.4 *Run the analysis*

7.4.1 Obtain measurements from all four collectors during each entry.

## 8. Calculation

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

8.1.1 For each entry, obtain a ratio by dividing the $UF_5^+$ ion count of the desired isotope by the $^{238}UF_5^+$ ion count.

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

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

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

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

$$(r_1 + r_6) / 2 = A \tag{1}$$

$$(r_2 + r_5) / 2 = X \tag{2}$$

$$(r_3 + r_4) / 2 = B \tag{3}$$

where $r_n$ is the ratio from the $n^{th}$ entry.

8.1.5 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 $^{238}U$.

$$E_A = \text{mass \%}\ ^{234}U / \text{mass \%}\ ^{238}U \tag{4}$$

$$H_A = \text{mass \%}\ ^{235}U / \text{mass \%}\ ^{238}U \tag{5}$$

$$Y_A = \text{mass \%}\ ^{236}U / \text{mass \%}\ ^{238}U \tag{6}$$

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

8.1.7 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 $Y_B - Y_A = D_Y$

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

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

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

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

NOTE 3—Calculations in 8.1.8 are based on an algebraic manipulation of Lagrange's interpolation formula using two known values of a function.

8.2 Calculate the mass percent as follows:

$$e = 100\ \% \cdot \mathrm{E_X} / (1 + \mathrm{E_X} + \mathrm{H_X} + \mathrm{Y_X}) \tag{10}$$

$$x = 100\ \% \cdot \mathrm{H_X} / (1 + \mathrm{E_X} + \mathrm{H_X} + \mathrm{Y_X}) \tag{11}$$

$$y = 100\ \% \cdot \mathrm{Y_X} / (1 + \mathrm{E_X} + \mathrm{H_X} + \mathrm{Y}) \tag{12}$$

where:

$e$ = mass % $^{234}U$,
$x$ = mass % $^{235}U$, and
$y$ = mass % $^{236}U$.

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

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

## 9. Precision and Bias

9.1 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 $^{234}$U, $^{235}$U, and $^{236}$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 $^{234}$U and $^{236}$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 $^{236}$U value assigned to G015-1. This value was obtained on a single-collector gas-phase mass spectrometer. The $^{235}$U reference values were assigned through a combination of thermal and gas-phase analysis.

9.2 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 3 mass spectrometers. Every run yielded 3 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 $^{234}$U, $^{235}$U, and $^{236}$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.

9.3 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. Pumping down the instrument and restarting the spectrometer by computer were the basic components of that variation.

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

| Identity | Function | $^{234}$U mass % | $^{235}$U mass % | $^{236}$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$^{A}$ |
| G015–1 | reference | 0.0060 | 1.5139 | 0.0152$^{A}$ |
| G020-1 | reference | 0.0156 | 2.0123 | 0.0039 |
| G025 | reference | 0.0204$^{A}$ | 2.4719 | 0.0018$^{B}$ |
| G030–1 | reference | 0.0224$^{A}$ | 3.0066 | 0.0040 |
| G030 | high std | 0.0186 | 3.0072 | 0.0108 |
| G050–1 | high std | 0.0264 | 4.9457 | 0.0369 |

$^{A}$ 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.

$^{B}$ 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 × % RSD is ± 2.2921 % for $^{234}$U, ± 0.0646 % for $^{235}$U, and ± 2.7263 % for $^{236}$U.

9.4 Due to the difficulties in movement and ownership of nuclear materials, as referred to in Section 1.4 of Guide C 1215, the reproducibility was obtained by treating the three spectrometers as three different laboratories. This should be taken into account when considering the reproducibility results.

9.5 *Precision*[3]—The precision was calculated based on mass percent units. The repeatability percent RSD for the $^{235}$U isotope was determined to be 0.006 % and was typically consistent for all levels of mass %. The reproducibility percent RSD for the $^{235}$U isotope was determined to be 0.010 % and tended to decrease as the assay increased.

9.5.1 The repeatability percent RSD for the $^{234}$U isotope was determined to be 0.411 % and typically decreased as the $^{234}$U mass percent increased. The reproducibility percent RSD for the $^{234}$U isotope was determined to be 0.722 %.

9.5.2 The average repeatability percent RSD for the $^{235}$U isotope was determined to be 0.344 % and tended to decrease as the $^{236}$U assay increased. The reproducibility percent RSD for the $^{236}$U isotope was determined to be 1.664 % and tended to decrease as the $^{236}$U assay increased. The precision results for the three uranium isotopes are listed in Table 2.

**TABLE 2 Precision and Bias Test Results in Mass Percent Units**

| Isotope | %Bias$^{A}$ | $S_r$$^{B}$ | %RSD$_r$$^{C}$ | $S_R$$^{D}$ | %RSD$_R$$^{E}$ |
|---|---|---|---|---|---|
| $^{235}$U | −0.003 | 0.00010 | 0.006 | 0.00014 | 0.010 |
| $^{234}$U | −0.810 | 0.000032 | 0.411 | 0.000085 | 0.722 |
| $^{236}$U | 3.711 | 0.000026 | 0.344 | 0.000120 | 1.664 |

$^{A}$ %Bias = (bias/reference value) × 100 %

$^{B}$ $S_r$= repeatability standard deviation

$^{C}$ %RSD$_r$= %RSD for repeatability

$^{D}$ $S_R$= reproducibility standard deviation

$^{E}$ %RSD$_R$= %RSD for reproducibility

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

9.6 *Bias*—The observed percent bias was calculated based on mass percent units. The observed percent bias for the $^{235}$U isotope was determined to be −0.003 %. The observed percent bias for the $^{234}$U isotope was determined to be −0.810 %. The observed percent bias for the $^{236}$U isotope was determined to be 3.711 %. The $^{235}$U and $^{234}$U data indicate no significant bias in the method. The $^{234}$U data indicate there may be a slight bias for that isotope. The bias results are listed in Table 2.

## 10. Keywords

10.1 double-standard; isotopic; mass spectrometer; multicollector; spectrometer; uranium hexafluoride

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[3] Supporting data have been filed at ASTM Headquarters and may be obtained by requesting Research Report RRxxx.xxxx.

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