



Standard Test Method for Isotopic Analysis of Uranium Hexafluoride by Double Standard Single-Collector Gas Mass Spectrometer Method¹

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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 with ^{235}U concentrations between 0.1 and 5.0 mass %.

1.2 This test method may be applicable for the entire range of ^{235}U concentrations for which adequate standards are available.²

1.3 This test method is for analysis by a gas magnetic sector mass spectrometer with a single collector using interpolation to determine the isotopic concentration of an unknown sample between two characterized UF_6 standards.

1.4 This test method is to replace the existing test method currently published in Test Methods C761 and is used in the nuclear fuel cycle for UF_6 isotopic analyses.

1.5 The values stated in SI units are to be regarded as standard. No other units of measurement are included in this standard.

1.6 *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:³

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

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 This test method applies to the measurement of ^{235}U . It would also be able to measure minor isotopes using the same principle subject to appropriate validation by the measurement laboratory.

³ 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.

C787 Specification for Uranium Hexafluoride for Enrichment

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

2.2 Other Document:

USEC-651 Uranium Hexafluoride: A Manual of Good Handling Practices, Latest Revisions

3. Summary of Test Method

3.1 The unknown sample and two standards, whose ^{235}U mass percent bracket that of the unknown, are introduced in sequence into the mass spectrometer and measurements are made that are a function of the mol ratio of ^{235}U to the total of the other isotopes of uranium. These measurements, together with the known composition of the standards, permit calculation of the ^{235}U composition of the sample by linear interpolation (1-6).

3.2 The symmetrical sequence of sample-standard introductions from low standard, sample, high standard, and then reversed is designed to minimize biases resulting from instrument drift, sample interaction or memory, and the nonlinearity of the relationship between the measured resistance ratios and the true sample mol ratios. Corrections generally are not required for instrument memory.

4. Significance and Use

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

4.2 ASTM Committee C26 Safeguards Statement:

4.2.1 The material (uranium hexafluoride) to which this test method applies is subject to the nuclear safeguards regulations governing its possession and use. The analytical procedure in this test method has been designated as technically acceptable for generating safeguards accountability data.

4.2.2 When used in conjunction with the appropriate certified reference materials (CRMs), this procedure can demonstrate traceability to the national measurement base. However, adherence to this procedure does not automatically guarantee

regulatory acceptance of the regulatory safeguards measurements. It remains the sole responsibility of the user of this test method to ensure that its application to safeguards has the approval of the proper regulatory authorities.

5. Apparatus

5.1 A mass spectrometer is used with the following features and capabilities (2-7):

5.1.1 An ion source with an accelerating voltage of approximately 1.75kv.

5.1.2 The sample inlet system shall have a minimum of three points for attachment of standards and samples, the necessary valves to evacuate the sample lines and admit the UF₆ into the ion source, and a variable leak to control the flow of UF₆ into the ion source.

5.1.3 A single ion collector (8) consisting of a high and low signal shall be used. The high-side current signal contains a central slit, preferably adjustable, to permit passage of greater than 90 % of the ions of the ²³⁵U isotope (mass 330). The ²³⁵U ions passing through the slit is called the low-side current signal.

5.1.4 The measuring system shall provide a mole ratio measurement calculation from the low- and high-current signal. Such a system usually consists of an amplifier and counter for each V-to-F convertor.

5.1.5 The resolving power of the instrument should be should be less than 5 % between Mass 330/333. This resolution requirement should be met with the collector slit width adjusted to pass at least 90 % of the ²³⁵U ion beam.

5.2 Computer control for opening and closing valves, timing sequence and integrations of signals and data acquisitions.

6. Procedure

6.1 Prepare Sample and Standards:

6.1.1 Select a high and low standard that bracket the ²³⁵U mass percent of the sample.

6.1.2 Attach sample and standard containers to the spectrometer.

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

6.1.4 Open the sample and standard containers individually to remove impurities that may bias the results or interfere with the ionization. If necessary, freeze the UF₆ with ice water or a mixture of crushed dry ice and isopropyl alcohol to permit longer venting without losing large amounts of UF₆.

6.1.5 Permit the exhaust system to recover.

6.1.6 Check to see if impurities have been sufficiently removed by introducing UF₆ into the ion source and observing pressure.

6.1.7 If necessary, repeat 6.1.4 to 6.1.6 until samples have been sufficiently purified.

6.2 Prepare Instrument:

6.2.1 Ensure the instrument parameters are set so the ²³⁵UF₅⁺ ion beam is focused through the slit on the high-side plate onto the low side plate.

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

6.2.3 For purposes of programming entry sequences into the spectrometer, let the low standard be represented by A, the unknown sample by X, and the high standard by B.

6.2.4 The most generic sequence to use in interpolating an unknown (X) between two standards is A, X, B or B, X, A. Combining an A, X, B with a B, X, A helps to eliminate bias as a result of residual amounts of UF₆ in the ion source from the previous entry. A sequence of A X B B X A is designed to do this. The data used in the precision and bias statement for this test method were obtained using this sequence. Other sequences of B X A A X B; A X B X A; or B X A X B are also designed to help alleviate this bias. Program the desired sequence.

6.2.5 Run the desired number of sequences for the analysis.

7. Calculation

7.1 For each entry, obtain a ratio of the low-side collector plate charge (²³⁵UF₅⁺) to the high-side collector plate charge (²³⁸UF₅⁺, ²³⁴UF₅⁺, and ²³⁶UF₅⁺).

7.2 For a sequence of A, X, B, B, X, A, find the arithmetic mean of the two ratios for A, X, and B. Let:

7.2.1 R_A = mean of the ratios for the low standard (A),

7.2.2 R_X = mean of the ratios for the sample (X), and

7.2.3 R_B = mean of the ratios for the high standard (B).

7.3 Calculate the ratio of differences, R_D, as follows:

$$R_D = (R_X - R_A)/(R_B - R_A) \quad (1)$$

7.4 Calculate the sample mass ratio, H_X, using linear interpolation as follows:

$$H_X = R_D (H_B - H_A) + H_A \quad (2)$$

where:

H_X = mass ratio of ²³⁵U to the other U isotopes in the sample,

H_A = mass ratio of ²³⁵U to the other U isotopes in the low standard, and

H_B = mass ratio of ²³⁵U to the other U isotopes in the high standard.

7.5 Calculate the mass % ²³⁵U in the sample as follows:

$$\text{Mass \% } ^{235}\text{U} = 100 H_X / (100 + H_X) \quad (3)$$

8. Precision and Bias

8.1 Three characterized UF₆ isotopic standards were run as control samples for this test method. All standards were traceable to recognized national standards. The standard designations, functions, and reference values are listed in Table 1.

8.2 Because of difficulties in movement and ownership of nuclear materials, the estimate of reproducibility was obtained

TABLE 1 Reference Materials and Their Assigned Values

Identity	Function	²³⁵ U Mass % Value ^A
ST 122-3	Depleted standard	0.52393
ST 128	Normal standard	0.71104
ST 26	Enriched standard	4.4601

^A The uncertainty in the assigned values, expressed as 3 × %RSD, is ±0.066 %.

by treating the spectrometers as the different laboratories. Each of the three standards was analyzed over several years by different analysts using different spectrometers in the same laboratory resulting in a total of 30 test results for each standard. The one analyst design was impractical because of the accumulation of data over such a large period of time. The contribution of variation as a result of different analysts was minimal. Pumping down the instrument and restarting the spectrometer by computer were the basic components of this variation.

8.3 For each reference material, the mean, repeatability standard deviation (SD_r), relative repeatability standard deviation ($\%RSD_r$), reproducibility standard deviation (SD_R), relative reproducibility standard deviation ($\%RSD_R$), repeatability limit, reproducibility limit, and % difference was calculated. The estimated precision results for the three characterized UF_6 isotopic standards are shown in [Table 2](#). Reproducibility results were obtained by treating each of the mass spectrometers as separate laboratories. This should be taken into account when considering the results.

8.4 *Precision*⁴—The precision was calculated based on mass percent units. The repeatability percent RSD (single spectrometer) was determined to be 0.017 % and was typically consistent for all levels of mass percent. The reproducibility percent RSD (between spectrometers) was calculated to be 0.018 %. The closeness in agreement between the repeatability and reproducibility values is expected since statistical analysis concluded no significant differences between mass spectrometer results.

8.5 *Bias*—The observed percent difference was calculated for each standard as an indication of bias and the estimates appear in [Table 2](#). The % difference (averaged over the three standards) was determined to be -0.009 % indicating an overall accuracy of 99.991 %.

9. Keywords

9.1 double standard; isotopic; mass spectrometry; single collector uranium hexafluoride

⁴ Supporting data have been filed at ASTM International Headquarters and may be obtained by requesting Research Report RR:C26-1013.

TABLE 2 Precision and Bias Test Results in ²³⁵U Mass Percent Units

Results	Reference Values			Average, %
	0.52393 ²³⁵ U Mass % Control	0.71104 ²³⁵ U Mass % Control	4.46010 ²³⁵ U Mass % Control	
Sample size	30	30	30	...
Mean	0.52391	0.71098	4.45950	...
SD _r ^A	0.000093	0.000147	0.00058	...
SD _R ^B	0.0001	0.000147	0.00058	...
%RSD _r ^C	0.018	0.021	0.013	0.017
%RSD _R ^D	0.019	0.021	0.013	0.018
Repeatability limit, % ^E	0.050	0.059	0.036	0.048
Reproducibility limit, % ^F	0.053	0.059	0.036	0.049
% Difference ^G	-0.0038	-0.0084	-0.0135	-0.009

^A SD_r = repeatability standard deviation (single spectrometer).

^B SD_R = reproducibility standard deviation (different spectrometers).

^C %RSD_r = relative repeatability standard deviation (percent) = 100 · (SD_r/mean).

^D %RSD_R = relative reproducibility standard deviation (percent) = 100 · (SD_R/mean).

^E Repeatability limit = 2.8 · %RSD_r. This represents the 95 % limits for the difference between two measurements taken by the same spectrometer.

^F Reproducibility limit = 2.8 · %RSD_R. This represents the 95 % limits for the difference between two measurements obtained by different spectrometers.

^G % Difference = 100 · [(mean – reference value)/reference value].

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