



Standard Test Method for Determination of Total Hydrogen Content of Uranium Oxide Powders and Pellets by Carrier Gas Extraction¹

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^{ε1} NOTE—Editorial corrections were made throughout in June 2010.

1. Scope

1.1 This test method applies to the determination of hydrogen in nuclear-grade uranium oxide powders and pellets to determine compliance with specifications. Gadolinium oxide (Gd_2O_3) and gadolinium oxide-uranium oxide powders and pellets may also be analyzed using this test method.

1.2 This standard describes a procedure for measuring the total hydrogen content of uranium oxides. The total hydrogen content results from absorbed water, water of crystallization, hydro-carbides and other hydrogenated compounds which may exist as fuel's impurities.

1.3 This test method covers the determination of 0.05 to 200 μg of residual hydrogen.

1.4 This test method describes an electrode furnace carrier gas combustion system equipped with a thermal conductivity detector.

1.5 The preferred system of units is micrograms hydrogen per gram of sample ($\mu g/g$ sample) or micrograms hydrogen per gram of uranium ($\mu g/g$ U).

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

1.7 *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:²

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 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.

[C753 Specification for Nuclear-Grade, Sinterable Uranium Dioxide Powder](#)

[C776 Specification for Sintered Uranium Dioxide Pellets](#)

[C888 Specification for Nuclear-Grade Gadolinium Oxide \(\$Gd_2O_3\$ \) Powder](#)

[C922 Specification for Sintered Gadolinium Oxide-Uranium Dioxide Pellets](#)

3. Summary of Test Method

3.1 The total hydrogen content is determined using a hydrogen analyzer. The hydrogen analyzer is based on the carrier gas method using argon or nitrogen as carrier gas. The actual configuration of the system may vary with vendor and model.

3.2 The samples to be analyzed are dropped into a preheated graphite crucible, and then, heated up to a temperature of more than 1700°C in a graphite crucible. At that temperature hydrogen, oxygen, nitrogen, and carbon monoxide (oxygen is converted to CO when it reacts with the crucible) are released. The release gas is purified in the carrier gas stream by oxidation and absorption columns. The hydrogen is separated by chromatographic means and analyzed in a thermal conductivity detector.

4. Significance and Use

4.1 Uranium dioxide is used as a nuclear-reactor fuel. Gadolinium oxide is used as an additive to uranium dioxide. In order to be suitable for this purpose, these materials must meet certain criteria for impurity content. This test method is designed to determine whether the hydrogen content meets Specifications [C753](#), [C776](#), [C888](#), and [C922](#).

5. Interferences

5.1 Contamination of carrier gas, crucibles, or samples with extraneous sources of hydrogen may cause a positive bias. A blank correction will help to minimize the bias from carrier gas and crucibles. Interference from adsorbed hydrogen on samples may be eliminated by keeping the sample in an inert atmosphere or vacuum.

5.2 The purification system typically associated with the recommended combustion and detection equipment is designed to minimize other expected sources of interferences, such as sulfur, halogens, carbon monoxide, carbon dioxide, and water.

5.2.1 The nitrogen and hydrogen peaks are close together and must be well-separated to prevent falsely high result from the nitrogen. The molecular sieve must be sufficiently long to separate the peaks and must be changed when the column becomes loaded with contaminants that prevent proper peak separation.

5.3 The temperature of >1700–1800°C must be reached. If not, the decomposition of the released water to hydrogen and carbon monoxide may not be complete. The temperature will depend upon the instrument and type of graphite crucible used. Single wall crucibles will require a lower temperature (power) than double wall crucibles.

5.4 Incomplete fusion may result in partial or a late release of hydrogen resulting in low results.

5.5 At temperatures of more than 2200°C uranium metal may be formed, and carbon dioxide released because of reduction of UO₂ by the graphite crucible.

5.5.1 Carbon dioxide will interfere with the thermal conductivity measurement. This interference can be minimized by use of chemical absorption, or a molecular sieve column, or both.

5.5.2 Excess temperature, from too much power, crucible hot spots, or from misaligned electrodes may cause analysis errors. Uranium samples should be evenly fused, fall out freely of the crucibles and contain very little uranium metal.

6. Apparatus

6.1 *Hydrogen Analyzer*, consisting of an electrode furnace capable of operation at least up to 2200 to 2500°C, a thermal conductivity detector for measuring, and auxiliary purification systems.

6.2 *Balance*, with precision of ± 1 mg.

7. Reagents and Materials

7.1 *Purity of Reagents*—Reagent grade chemicals shall be used in all tests. Unless otherwise indicated, it is intended that all reagents shall conform to the specifications of the Committee on Analytical Reagents of the American Chemical Society, where such specifications are available. Other grades may be used, provided it is first ascertained that the reagent is of sufficiently high purity to permit its use without lessening the accuracy of the determination.

7.2 *Carrier Gas*—Nitrogen $\geq 99.998\%$ or Argon $\geq 99.995\%$.

7.3 *Carrier Gas Purifiers*:

7.3.1 *Copper Oxide*, or rare earth copper oxide (converts H to H₂O), or

7.3.2 *Copper Turnings*, or granules.

7.4 *Molecular Sieve-Sodium Hydroxide*, on a fiber support (sodium hydroxide reacts with CO₂ to yield water; the molecular sieve separates N₂ and H₂).

7.5 *Schutze Reagent*, iodine pentoxide over silica gel (converts CO to CO₂).

7.6 *Magnesium Perchlorate*—removes water.

7.7 *Silicone Vacuum Grease*.

7.8 *Tin Flux*, if Zr or Ti hydride standards are to be used.

7.9 *Graphite Crucibles*.

7.10 *Tin Capsules*.

7.11 *Aluminum Oxide (Al₂O₃)*, to check furnace temperature.

7.12 *Hydrogen Standard Materials*—Calibrate the instrument using either high purity (99.9999 %) certified hydrogen gas or NIST-traceable, or equivalent, metal standards. Steel standards³ are the preferred metal standards because no flux is used, and this best matches the conditions used to analyze uranium oxide samples. Zr- or Ti-hydride standards may be used, but require the use of a flux metal.

7.13 *Sodium Tartrate or Sodium Tungstate* may be used as check standards for uranium powder analyses.

8. Hazards and Precautions

8.1 Take proper safety precautions to prevent inhalation or ingestion of uranium dioxide powders or dust during grinding or handling operations.

8.2 Operation of equipment presents electrical and thermal hazards. Follow the manufacturer's recommendations for safe operation.

8.3 This procedure uses hazardous chemicals. Use appropriate precautions for handling corrosives, oxidizers, and gases.

9. Preparation of Apparatus

9.1 Inspect and change instrument column packing and reagents as recommended by manufacturer.

9.2 Check to ensure that the furnace heats properly on a periodic basis. A quarterly check is recommended. A properly functioning furnace, set at normal operating parameters should fuse Al₂O₃ (approximately 2050°C melting point, depending upon form).

9.3 Set the operating controls of the instrument system according to the operating instructions for the specific equipment used.

9.4 Condition the apparatus by combustion of several blanks prepared with sample crucible and accelerator, if any, in the amount to be used with the samples. Successive blanks should approach a constant value, allowing for normal statistical fluctuations.

9.5 The blank measurements prove the integrity of the purifying units and the tightness of the equipment. Blank values of more than ± 0.03 $\mu\text{g H}_2$ require adequate measures of correction.

10. Calibration Using Metal Standards

10.1 The calibration range and number of standards will depend upon the instrument used. Three to five standards, containing 3 to 6 μg hydrogen are recommended. The number of standards and calibration range will depend upon the availability, assay accuracy, and homogeneity of available standards.

³ NIST-traceable steel standards marketed by LECO have been found to perform satisfactorily.