



# Standard Test Methods for Chemical, Mass Spectrometric, and Spectrochemical Analysis of Nuclear-Grade Uranium Dioxide Powders and Pellets<sup>1</sup>

This standard is issued under the fixed designation C 696; 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 These test methods cover procedures for the chemical, mass spectrometric, and spectrochemical analysis of nuclear-grade uranium dioxide powders and pellets to determine compliance with specifications.

1.2 The analytical procedures appear in the following order:

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C 1413 Test Method for Isotopic Analysis of Hydrolysed Uranium Hexafluoride and Uranyl Nitrate Solutions By Thermal Ionization Mass Spectrometry

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## 2. Referenced Documents

### 2.1 ASTM Standards:

- C 753 Specification for Nuclear-Grade, Sinterable Uranium Dioxide Powder<sup>3</sup>
- C 761 Test Method for Chemical, Mass Spectrometric, Spectrochemical, Nuclear, and Radiochemical Analysis of Uranium Hexafluoride<sup>3</sup>
- C 776 Specification for Sintered Uranium Dioxide Pellets<sup>3</sup>
- C 1267 Test Method for Uranium By Iron (II) Reduction In Phosphoric Acid Followed By Chromium (VI) Titration In The Presence of Vanadium<sup>3</sup>
- C 1287 Test Method for Determination of Impurities In Uranium Dioxide By Inductively Coupled Plasma Mass Spectrometry<sup>3</sup>
- C 1347 Practice for Preparation and Dissolution of Uranium Materials for Analysis<sup>3</sup>
- C 1408 Test Method for Carbon (Total) in Uranium Oxide Powders and Pellets By Direct Combustion-Infrared Detection Method<sup>3</sup>
- C 1413 Test Method for Isotopic Analysis of Hydrolysed Uranium Hexafluoride and Uranyl Nitrate Solutions By Thermal Ionization Mass Spectrometry<sup>3</sup>
- D 1193 Specification for Reagent Water<sup>5</sup>
- E 115 Practice for Photographic Processing in Optical Emission Spectrographic Analysis<sup>6</sup>
- E 116 Practice for Photographic Photometry by Spectrochemical Analysis<sup>6</sup>
- E 130 Practice for Designation of Shapes and Sizes of Graphite Electrodes<sup>6</sup>
- E 217 Test Method for Uranium by Controlled-Potential Coulometry<sup>3</sup>
- E 305 Practice for Establishing and Controlling Spectrochemical Analytical Curves<sup>6</sup>

<sup>1</sup> These test methods are under the jurisdiction of ASTM Committee C-26 on Nuclear Fuel Cycle.

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<sup>2</sup> Discontinued January 1999. See C 696–80.

<sup>3</sup> *Annual Book of ASTM Standards*, Vol 12.01.

<sup>4</sup> Discontinued as of May 30, 1980.

<sup>5</sup> *Annual Book of ASTM Standards*, Vol 11.01.

<sup>6</sup> *Annual Book of ASTM Standards*, Vol 03.05.

E 402 Test Method for Spectrographic Analysis of Uranium Oxide ( $U_3O_8$ ) by Gallium Oxide-Carrier Technique<sup>7</sup>

### 3. Significance and Use

3.1 Uranium dioxide is used as a nuclear-reactor fuel. In order to be suitable for this purpose, the material must meet certain criteria for uranium content, stoichiometry, isotopic composition, and impurity content. These test methods are designed to show whether or not a given material meets the specifications for these items as described in Specifications C 753 and C 776.

3.1.1 An assay is performed to determine whether the material has the minimum uranium content specified on a dry weight basis.

3.1.2 The stoichiometry of the oxide is useful for predicting its sintering behavior in the pellet production process.

3.1.3 Determination of the isotopic content of the uranium in the uranium dioxide powder is made to establish whether the effective fissile content is in compliance with the purchaser's specifications.

3.1.4 Impurity content is determined to ensure that the maximum concentration limit of certain impurity elements is not exceeded. Determination of impurities is also required for calculation of the equivalent boron content (EBC).

### 4. Reagents

4.1 *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.<sup>8</sup> 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.

4.2 *Purity of Water*— Unless otherwise indicated, references to water shall be understood to mean reagent water conforming to Specification D 1193.

### 5. Safety Precautions

5.1 Proper precautions should be taken to prevent inhalation, or ingestion of uranium dioxide powders or dust during grinding or handling operations.

### 6. Sampling

6.1 Criteria for sampling this material are given in Specification C 753 and Specification C 776.

6.2 Samples can be dissolved using the appropriate dissolution techniques described in Practice C 1347, but final determination of applicability must be made by the user.

### URANIUM BY FERROUS SULFATE REDUCTION IN PHOSPHORIC ACID AND DICHROMATE TITRATION METHOD

This Test method was discontinued in January 1999 and replaced by Test method C 1267.

### URANIUM AND OXYGEN TO URANIUM ATOMIC RATIO BY THE IGNITION (GRAVIMETRIC) IMPURITY CORRECTION METHOD

#### 7. Scope

7.1 This test method covers the determination of uranium and the oxygen to uranium atomic ratio in nuclear-grade uranium dioxide powder and pellets.

#### 8. Summary of Test Method

8.1 A weighed portion of  $UO_2$  is dried under reduced pressure in a nitrogen atmosphere, desiccated, and weighed. The dried oxide is then converted to  $U_3O_8$  by ignition at  $900^\circ C$  (8, 9).

#### 9. Interferences

9.1 The weight of  $U_3O_8$  is corrected for the nonvolatile impurities present as determined by spectrographic analysis. An extended ignition time may be required if significant amounts of anions that are difficult to decompose are present.

#### 10. Apparatus

10.1 *Vacuum Oven*, capable of maintaining and controlling temperatures to  $180^\circ C$  and equipped with double stopcocks and a vacuum gage (range from 0 to 102 kPa (0 to 30 in. Hg)).

10.2 *Drying Tower*— Prepare a U-tube filled with a carbon dioxide absorbent<sup>9</sup> and a suitable moisture absorbent, that is, anhydrous magnesium perchlorate  $Mg(ClO_4)_2$ .

10.3 *Muffle Furnace*, capable of maintaining and controlling temperatures to  $1000^\circ C$ .

#### 11. Procedure

11.1 Transfer approximately 5 to 10 g of  $UO_2$  powder or up to 50 g of pellets to a tared platinum crucible and weigh to within 0.1 mg.

11.2 Place the crucible in a vacuum oven set at room temperature, seal the oven, and reduce the pressure to approximately 95 to 102 kPa (28 to 30 in. Hg).

11.3 Close the vacuum valve and slowly flush the oven with dry nitrogen.

11.4 Close the nitrogen inlet and reduce the pressure to 95 to 102 kPa (28 to 30 in. Hg). Repeat the nitrogen flush as in step 11.3 to give a total of three flushes.

11.5 Close the nitrogen inlet valve, reduce the pressure to 95 to 102 kPa (28 to 30 in. Hg), set the temperature at  $45^\circ C$  for powder samples or  $160^\circ C$  for pellets, and maintain these conditions for 4 h. After 4 h of heating turn off the heat and allow the oven to cool to room temperature while under reduced pressure.

11.6 Turn off the vacuum valve and slowly introduce dry nitrogen until the oven door can be opened.

<sup>7</sup> Annual Book of ASTM Standards, Vol 03.06.

<sup>8</sup> Reagent Chemicals, American Chemical Society Specifications, American Chemical Society, Washington, DC. For suggestions on the testing of reagents not listed by the American Chemical Society, see *Analar Standards for Laboratory Chemicals*, BDH Ltd., Poole, Dorset, U.K., and the *United States Pharmacopeia and National Formulary*, U.S. Pharmaceutical Convention, Inc. (USPC), Rockville, MD.

<sup>9</sup> Ascarite has been found satisfactory for this purpose.

11.7 Transfer the crucible to a desiccator and cool. Remove the crucible and weigh immediately (8).

11.8 Place the crucible containing the dried oxide in a muffle furnace set at 900°C. Ignite powder samples for 3 h. Pellets should be preheated at 500°C for 3 h, then ignited 3 h at 900°C.

11.9 Remove the crucible from the furnace, allow to cool in the air 2 to 3 min then place the crucible in a desiccator and cool to room temperature. Weigh the crucible and repeat steps 11.8 and 11.9 until a constant weight is obtained.

11.10 Submit the ignited sample for spectrographic analysis.

## 12. Calculation

12.1 *Loss on Vacuum Drying*—Calculate as follows based on original sample:

$$\text{Loss, wt \%} = [(S - W_1)/S] \times 100 \quad (1)$$

where:

$S$  = initial sample mass, and

$W_1$  = sample mass after vacuum drying, g.

12.2 *Uranium Content*— Calculate as follows:

$$U, \text{ wt \%} = [(0.8480 (W_2 - W_2I)/S) \times 100] - 0.01 \% \text{ (Note 1)} \quad (2)$$

where:

0.8480 =  $U_3O_8$  to uranium conversion factor for natural uranium. Corrections must be made in this factor as the uranium isotopic abundance deviates from natural uranium,

$W_2$  = grams of  $U_3O_8$  after ignition,

$S$  = initial sample mass, and

$I$  = total grams of all impurity-element oxides per gram of ignited  $U_3O_8$  (Note 2).

NOTE 1—All nonvolatile impurity values reported as less than the threshold of detection are considered to contribute a total correction of 0.01 % to the uranium percent.

NOTE 2—See Table 1 to obtain conversion factors for many common impurity elements encountered.

12.3 *Oxygen-to-Uranium Ratio*—Calculate as follows from the original sample,  $U$  wt %:

$$O/U = [(100 - U \text{ wt \%} - Z - n)(A)][15.999(U) \text{ wt \%}] \quad (3)$$

where:

$A$  = atomic mass of uranium based on isotopic abundance,

$O$  = atom % of oxygen,

$U$  = atom % of uranium,

$n$  = moisture content, %, and

$Z$  = total impurities correction, %.

## 13. Precision and Accuracy

13.1 For atomic ratios of O/U in the range from 2.00 to 2.10 the standard deviation was found to be 0.007 absolute at 95 % confidence level.

TABLE 1 Conversion Factors for Impurity Correction

Impurity	Assumed Oxide Form	Gravimetric Factor
Al	Al <sub>2</sub> O <sub>3</sub>	1.89
Ba	BaO	1.12
Be	BeO	2.78
Bi	Bi <sub>2</sub> O <sub>3</sub>	1.11
Ca	CaO	1.40
Cd	CdO	1.14
Co	Co <sub>2</sub> O <sub>3</sub>	1.41
Cr	Cr <sub>2</sub> O <sub>3</sub>	1.46
Cu	CuO	1.25
Fe	Fe <sub>2</sub> O <sub>3</sub>	1.43
Li	Li <sub>2</sub> O	2.15
Mg	MgO	1.66
Mn	MnO <sub>2</sub>	1.58
Mo	MoO <sub>3</sub>	1.50
Na	Na <sub>2</sub> O	1.35
Ni	NiO	1.27
P	P <sub>2</sub> O <sub>5</sub>	2.29
Pb	PbO <sub>2</sub>	1.15
Sb	Sb <sub>2</sub> O <sub>4</sub>	1.26
Si	SiO <sub>2</sub>	2.14
Sn	SnO <sub>2</sub>	1.27
Ti	TiO <sub>2</sub>	1.67
V	V <sub>2</sub> O <sub>5</sub>	1.79
Zn	ZnO	1.24
Zr	ZrO <sub>2</sub>	1.35
Ta	Ta <sub>2</sub> O <sub>5</sub>	1.22
W	WO <sub>3</sub>	1.26

### CARBON (TOTAL) BY DIRECT COMBUSTION-THERMAL CONDUCTIVITY METHOD

This Test Method was discontinued in January 1999 and replaced by Test Method C 1408

### TOTAL CHLORINE AND FLUORINE BY PYROHYDROLYSIS ION-SELECTIVE ELECTRODE METHOD

## 14. Scope

14.1 This test method covers the determination of chlorine and fluorine in nuclear-grade uranium dioxide. With a 1 to 10-g sample, concentrations of 5 to 200 µg/g of chlorine and 1 to 200 µg/g of fluorine are determined without interference.

## 15. Summary of Test Method

15.1 The halogens are separated from powdered uranium dioxide by pyrohydrolysis in a quartz tube with a stream of wet oxygen at a temperature of 900 to 1000°C (10, 11, 12, 13). Chloride and fluoride are volatilized as acids, absorbed in a buffer solution, and measured with ion-selective electrodes (13, 14, 15).

## 16. Apparatus

16.1 *Pyrohydrolysis Equipment*—A suitable assembly of apparatus is shown in Fig. 1.

16.1.1 *Gas Flow Regulator and Flowmeter*.

16.1.2 *Hot Plate*, used to warm the water saturating the sparge gas to 50 to 80°C.

16.1.3 *Combustion Tube Furnace*, having a bore of about 32 mm (1¼in.), a length of about 305 mm (12 in.), and the capability of maintaining a temperature of 1000°C.

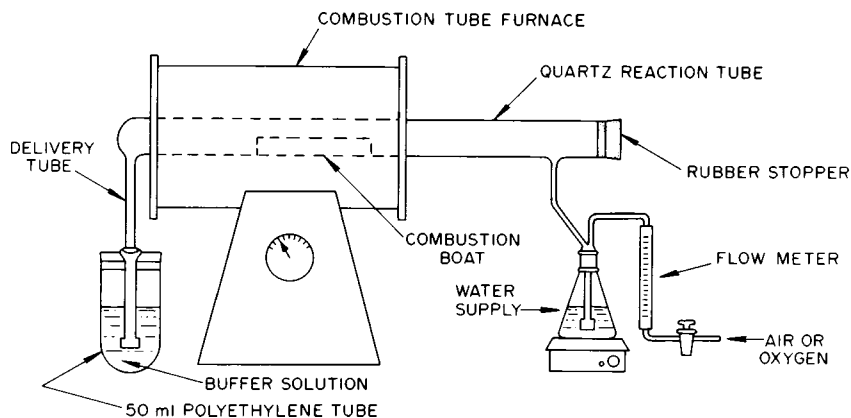


FIG. 1 Pyrohydrolysis Apparatus

16.1.4 *Quartz Reaction Tube* (Fig. 2)—The exit end should not extend over 51 mm (2 in.) beyond the furnace with a ground joint connecting to the delivery tube. The delivery tube extends into a polyethylene absorption vessel with a tip capable of giving a stream of fine bubbles.

16.1.5 *Combustion Boat*—A platinum or quartz boat with a 10-ml capacity (89 to 102 mm (3½ to 4 in.) long, 12.7 mm (½ in.) wide, and 9.53 mm (⅜ in.) high).

16.1.6 *Absorption Vessel*—A 50-mL polyethylene graduate or tube is satisfactory.

16.2 *Ion-Specific Electrodes*—A fluoride-specific activity electrode<sup>10</sup>; chloride-specific electrode.<sup>11</sup>

16.3 *pH Meter and Double-Junction Reference Electrode*, such as a mercuric sulfate, sleeve junction type. The meter should have an expandable scale with a sensitivity of 1 mV.

16.4 *Magnetic Stirrer*.

16.5 *Beakers*, 50-mL polyethylene.

17. Reagents

17.1 *Accelerator, U<sub>3</sub>O<sub>8</sub> halogen-free*, can be used but a flux of sodium tungstate (Na<sub>2</sub>WO<sub>4</sub>) with tungsten trioxide (WO<sub>3</sub>) may be used to advantage (10, 11). Special preparation of the mixture is necessary.

NOTE 3—Dehydrate 165 g of Na<sub>2</sub>WO<sub>4</sub> in a large platinum dish. Transfer the dried material to a mortar, add 116 g of WO<sub>3</sub>, and grind the mixture to ensure good mixing. Transfer the mixture into a platinum dish

<sup>10</sup> The Orion Model 9409 has been found satisfactory.

<sup>11</sup> The Orion Model No. 96-17-00 has been found satisfactory.

and heat with a burner for 2 h. Cool the melt, transfer the flux to a mortar and grind to a coarse powder. Store the flux in an airtight bottle. Mix about 8 g of flux with each portion of sample to be pyrohydrolyzed.

17.2 *Buffer Solution*— Dissolve 0.1 g potassium acetate (KC<sub>2</sub>H<sub>3</sub>O<sub>2</sub>) in water, add 0.050 mL of acetic acid (CH<sub>3</sub>CO<sub>2</sub>H, sp gr 1.05), and dilute to 1 litre.

17.3 *Chloride, Standard Solution (1 mL = 100 µg Cl)*— Dissolve 165 mg of dry sodium chloride (NaCl) in water and dilute to 1 litre.

17.4 *Distilled Water*— The water must be free of all chlorides and fluorides.

17.5 *Fluoride, Standard Solution (1 mL = 50 µg F)*— Dissolve 111 mg of sodium fluoride (NaF) in water and dilute to 1 litre. Store the solution in a polyethylene bottle.

17.6 *Compressed Oxygen or Air*.

18. Procedure

18.1 Adjust the pyrohydrolysis system to operating condition as follows:

- 18.1.1 Heat the furnace to 950 ± 50°C.
- 18.1.2 Fill the water reservoir, and heat to 50 to 80°C.
- 18.1.3 Adjust the gas flow to about 1.5 to 2 litres/min.

18.2 Flush the reaction tube and boat with moist oxygen or according to the pyrohydrolysis procedure in 18.4.

18.3 Run a pyrohydrolysis blank using a halide-free uranium oxide according to the procedure in 18.4. A blank run should be made each day.

18.4 *Sample Pyrohydrolysis:*

18.4.1 Weigh 1 to 5 g of powdered UO<sub>2</sub> and spread in the combustion boat. If an accelerator is desired, mix 4 g of U<sub>3</sub>O

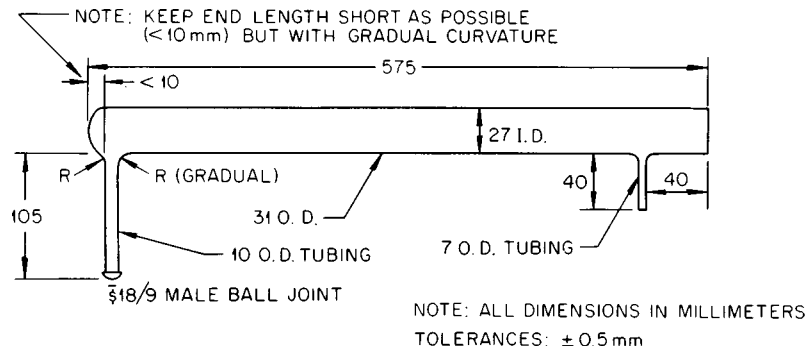


FIG. 2 Quartz Reaction Tube

8 or 8 g of the tungstate flux with the  $\text{UO}_2$  before spreading in the boat.

18.4.2 Place 15 mL of acetate buffer solution in the collection flask and submerge the delivery tip in the solution.

18.4.3 Remove the stopper from the entrance of the reaction tube and insert the boat into the hot area of the furnace. Restopper the furnace tube.

18.4.4 Check the gas flow and adjust to 1.5 to 2 litres/min.

18.4.5 Continue the reaction for 1 h. Thirty minutes may be sufficient with the tungstate flux.

18.4.6 To establish the time required for complete pyrohydrolysis, replace the buffer solution and continue the reaction for an additional 30 min.

18.4.7 When the pyrohydrolysis is completed, the buffer solution is transferred to a 25-mL flask. Rinse the delivery tube and collection tube with a minimum of buffer solution. Make up to volume. Use 10-mL aliquots of the diluted condensate for each determination.

#### 18.5 Chloride and Fluoride Measurement:

18.5.1 Assemble the meter and electrode in accordance with the instructions with the ion-specific electrode and the expanded scale meter being used.

18.5.2 Use successive dilutions of the chloride and fluoride standards in the buffer solution on a 25-mL volume basis to prepare calibration curves for each electrode. Plot the millivolt readings of a series of standards versus the concentration in micrograms per 25 mL on semi-log paper. The concentration of chloride should cover 10  $\mu\text{g}/25$  mL to 100  $\mu\text{g}/25$  mL and the fluoride from 5  $\mu\text{g}/25$  mL to 100  $\mu\text{g}/25$  mL.

## 19. Calculation

19.1 Chlorine—Calculate as follows:

$$\text{Cl, } \mu\text{g/g} = (C - B)/W \quad (4)$$

where:

$C$  = micrograms of total chlorine in absorber solution,

$B$  = micrograms of total chlorine in the pyrohydrolysis blank, and

$W$  = grams of  $\text{UO}_2$  specimen pyrohydrolyzed

19.2 Fluorine—Calculate as follows:

$$\text{F, } \mu\text{g/g} = (F - B)/W \quad (5)$$

where:

$F$  = micrograms of total fluorine in absorber solution,

$B$  = micrograms of total fluorine in the pyrohydrolysis blank, and

$W$  = grams of  $\text{UO}_2$  specimen pyrohydrolyzed.

## 20. Precision

20.1 Recovery test runs with spiked oxide samples indicate that a precision of at least 10 % can be expected.

## MOISTURE BY THE COULOMETRIC ELECTROLYTIC MOISTURE ANALYZER METHOD

### 21. Scope

21.1 This test method covers the determination of moisture in uranium dioxide samples. Detection limits are as low as 10  $\mu\text{g}$ .

## 22. Summary of Test Method

22.1 The sample is heated in an oven (up to 400°C) to drive off any water. The moisture is carried from the oven into the electrolytic cell by a flowing stream of dry nitrogen. Two parallel platinum wires wound in a helix are attached to the inner surface of the tube, the wall of which is evenly coated with phosphorus pentoxide ( $\text{P}_2\text{O}_5$ ) (a strong desiccant that becomes electrically conductive when wet). A potential applied to the wires produces a measurable electrolysis current when moisture wets the desiccant. Electrolysis of the water continuously regenerates the cell enabling it to accept additional water.

22.2 Precautions must be taken to prevent interference from the following sources. Hydrogen fluoride will cause permanent damage to the cell and sample system and should not be run under any conditions. Corrosive acidic gases, such as chlorine and hydrogen chloride, will corrode the instrument. Entrained liquids and solids can cause cell failure and should be prevented from entering the gas stream. Ammonia and other basic materials react with the acidic cell coating and renders the cell unresponsive. Hydrogen, and to a lesser extent, oxygen or air, may cause a high reading due to recombination, in the cell, or in the case of hydrogen, due to reaction with oxide coating of the sample boat to produce water. Alcohols and glycols, particularly the more volatile ones, respond like water and therefore must not be present.

## 23. Apparatus

23.1 Moisture Analyzer, for solids, with quartz glass oven capable of being heated from ambient temperatures to 1000°C. The assembly includes electrolytic cell, flow meter, range 30 to 140  $\text{cm}^3/\text{min}$  air, and a dryer assembly.<sup>12</sup>

23.2 Balance,<sup>13</sup> for weighing samples in the range from 1 to 100 mg.

23.3 Nitrogen Gas Cylinder, with a pressure regulator, a flow meter and a drying tower.

## 24. Reagents

24.1 Barium Chloride Dihydrate ( $\text{BaCl}_2 \cdot 2 \text{H}_2\text{O}$ ).

## 25. Operation

25.1 Turn the main power switch ON.

25.2 Adjust nitrogen gas pressure to 41.4 kPa (6 psi) and the flow rate to 50 mL/min measured at the exit of the apparatus.

25.3 Weigh the sample into a small, dry, aluminum boat (Note 4) and insert it into the instrument oven as follows:

NOTE 4—For samples that have been reduced in a hydrogen atmosphere and thus contain excess hydrogen, the use of a platinum boat in place of the aluminum tube and nickel boat will minimize any interference due to the hydrogen.

25.3.1 Open the top of the analyzer and remove the TFE-fluorocarbon plug. Do not touch with gloves.

25.3.2 With forceps pull the nickel boat one third of the way out of the tube and place the aluminum boat and the sample

<sup>12</sup> A CEC Solids Moisture Analyzer, of Type 26-321A-MA is available from DuPont Instruments Inc., S. Shamrock Ave., Monrovia, CA 91016.

<sup>13</sup> A Cahn Electrobalance, or equivalent, available from Cahn Division, Ventum Instrument Corp., Paramount, CA has been found satisfactory.

inside the nickel boat, then reposition the nickel boat near the center of the heating coils.

25.3.3 Replace the TFE-fluorocarbon plug and close the lid of the analyzer.

25.4 Reset the counter to 0  $\mu\text{g}$ .

25.5 Set the timer at 1 h.

25.6 Set the temperature at 400°C. This will activate the analyzer and start the heating cycle.

25.7 When the preset temperature has been reached and the counter ceases counting, record the reading, *S*.

## 26. Standardization

26.1 Determine the blank by processing dry, empty, aluminum boats according to steps 25.3-25.7 until constant values are obtained.

26.2 Weigh and analyze replicate 5-mg samples of  $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$  until consistent results are obtained. Sodium tungstate dihydrate ( $\text{Na}_2\text{WO}_4 \cdot 2\text{H}_2\text{O}$ ) may also be used for calibration.

## 27. Calculation

27.1 Calculate the moisture recovery, *Z*, for the standard as follows:

$$Z = (A - B)147.2Y \quad (6)$$

where:

*A* = micrograms of moisture on counter when standard is tested,

*B* = micrograms of moisture on counter from blank, and  
*Y* = milligrams of  $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$ . Each milligram of  $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$  contains 147.2  $\mu\text{g}$  of water.

27.2 Calculate the percent moisture in the sample as follows:

$$\text{Moisture, \%} = [(S - B)/1000 WZ] \times 100 = (S - B)/10 WZ \quad (7)$$

where:

*S* = micrograms of moisture on counter when sample is tested,

*B* = micrograms of moisture on counter from blank,

*W* = milligrams of sample, and

*Z* = recovery of moisture from standard.

## 28. Precision

28.1 The relative standard deviation for moisture in a concentration range of 100  $\mu\text{g/g}$  is approximately 2 % but increases to 10 % at the 20  $\mu\text{g/g}$  level.

## NITROGEN BY THE KJELDAHL METHOD

### 29. Scope

29.1 This test method covers the determination of nitride nitrogen in uranium dioxide in the range from 10 to 250  $\mu\text{g}$ .

### 30. Summary of Test Method

30.1 The sample is decomposed with acid, the resulting solution is made strongly alkaline with sodium hydroxide solution, and the nitrogen is separated as ammonia by steam distillation. The distillate is collected in boric acid solution and the ammonia present is titrated with 0.01 *N* standard acid using a mixed indicator.

NOTE 5—Although a simple acid digestion is usually adequate for dissolution of uranium samples, some uranium nitrides do not yield to such treatment. The use of potassium dichromate in phosphoric acid (16) has proved to be successful with nitrides that are difficult to decompose. Therefore, this medium has been recommended although, in most cases, a mixture of phosphoric and sulfuric acids would be adequate.

### 31. Interferences

31.1 There should be no interferences in nuclear-grade uranium dioxide.

### 32. Apparatus

32.1 *Nitrogen Distillation Apparatus*, micro.<sup>14</sup>

32.2 *Heater*, 750-W electric, full-control.

32.3 *Burner*, bunsen-type.

32.4 *Buret*, micro, class A, 5 or 10-mL capacity, graduated in 0.02-mL divisions.

### 33. Reagents

33.1 *Ammonia-Free Water*—Prepare by distillation or from an ion-exchange column.

33.2 *Boric Acid-Indicator Solution*—Dissolve 20 g of boric acid ( $\text{H}_3\text{BO}_3$ ) in 800 mL of hot ammonia-free water, cool the solution, add 4 mL of mixed indicator solution (52.3), and dilute to 1 litre.

33.3 *Mixed Indicator Solution*—Mix 100 mL of a 1 % alcoholic solution of bromocresol green and 20 mL of a 1 % alcoholic solution of methyl red.

33.4 *Phosphoric Acid* ( $\text{H}_3\text{PO}_4$ , 85 %)—Heat acid to 190°C to remove excess water.

NOTE 6—Some lots of  $\text{H}_3\text{PO}_4$  give high blanks and cannot be used.

33.5 *Potassium Dichromate Solution (65 g/litre)*—Dissolve 65 g of potassium dichromate ( $\text{K}_2\text{Cr}_2\text{O}_7$ ) in ammonia-free water and dilute to 1 litre. If necessary to reduce the blanks prepare the dichromate by recrystallization of  $\text{K}_2\text{CrO}_4$  from alkaline solution (16).

33.6 *Sodium Hydroxide Solution*—Dissolve 500 g of sodium hydroxide ( $\text{NaOH}$ ) in 1 litre of ammonia-free water.

33.7 *Sulfuric Acid, Standard*—( $\text{H}_2\text{SO}_4$ , 0.01 *N*)—Standardize against a standard sodium hydroxide solution that has been standardized against potassium hydrogen phthalate.

NOTE 7—Hydrochloric acid ( $\text{HCl}$ , 0.01 *N*) may be used instead of  $\text{H}_2\text{SO}_4$ .

### 34. Procedure

34.1 *Blank Determinations*:

34.1.1 Fill the boiler of the distillation apparatus with ammonia-free water and distill for at least 30 min with a digestion flask in place in order to purge the apparatus of any traces of ammonia present.

34.1.2 Place 10 mL of  $\text{H}_3\text{PO}_4$  and 15 mL of potassium dichromate solution (65 g/litre) in a digestion flask and attach to the apparatus. Add 50 mL of  $\text{NaOH}$  solution and start passing the steam from the boiler through the digestion flask.

34.1.3 Place a 125-mL Erlenmeyer flask containing 5 mL of the boric acid-indicator solution over the tip of the condenser

<sup>14</sup> Kemmerer-Hallett Type, Fisher Scientific Co., has been found satisfactory.

and collect 25 mL of distillate. Lower the flask so that the tip of the condenser is above the level of the distillate and continue the distillation for an additional 30 s to rinse down the inside of the tube.

34.1.4 Titrate the distillate with the 0.01 N H<sub>2</sub>SO<sub>4</sub> from a microburet until the solution turns to a pink color.

34.1.5 Repeat the blank determination, steps 34.1.2-34.1.4, until the blanks are constant. If the blank exceeds 0.03 to 0.04 mL, look for a source of contamination.

#### 34.2 Analysis of the Sample:

34.2.1 Transfer up to 2 g of a weighed, powdered sample (Note 8) to the digestion flask.

NOTE 8—Samples in pellet form must be crushed in a diamond mortar to - 100 mesh powder and sampled by riffling or quartering to obtain a representative sample.

34.2.2 Add 10 mL of H<sub>3</sub>PO<sub>4</sub> and heat the flask gently with a small burner until a clear green solution is obtained. Inspect the solution carefully to ensure that no undissolved uranium nitrides remain.

34.2.3 Cool the flask, then add 15 mL of K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub> solution (65 g/litre) slowly with mixing. Warm at low heat for 3 to 4 min.

34.2.4 Attach the digestion flask to the distillation apparatus and add 50 mL of NaOH solution.

34.2.5 Place the receiving flask containing 5 mL of the boric acid-indicator solution over the condenser tip and distill and titrate following the procedure used to determine the blank.

### 35. Calculation

35.1 Calculate the nitrogen content as follows:

$$N, \mu\text{g/g on UO}_2 \text{ basis} = (A - B) 14.01 N \times 10^3 / W \quad (8)$$

where:

- A = millilitres of standard acid to titrate sample,
- B = millilitres of standard acid to titrate blank,
- N = normality of standard acid solution, and
- W = grams of UO<sub>2</sub> sample.

### 36. Precision

36.1 This test method will determine nitrogen to within 7  $\mu\text{g}$  of the amount present.

#### ISOTOPIC URANIUM COMPOSITION BY MULTIPLE-FILAMENT SURFACE-IONIZATION MASS SPECTROMETRIC METHOD

(This test method was discontinued in 1980 and replaced by Test Method C 1413.)

#### SPECTROCHEMICAL DETERMINATION OF TRACE ELEMENTS IN HIGH-PURITY URANIUM DIOXIDE

### 37. Scope

37.1 This test method covers the spectrographic analysis of nuclear-grade UO<sub>2</sub> for the 26 elements in the ranges indicated in Table 2.

37.2 For simultaneous determination of trace elements by plasma emission spectroscopy refer to Test Method C 761.

### 38. Summary of Test Method

38.1 The sample of UO<sub>2</sub> is converted to U<sub>3</sub>O<sub>8</sub> and mixed

TABLE 2 Recommended Analytical Spectral Lines and Concentration Range of Trace Elements

Element	Analytical Line, °A <sup>A</sup>	Concentration range, $\mu\text{g/g}$ of U
Ag <sup>B</sup>	3280.68	0.1 to 50
Al	2367.06	10 to 200
As	2349.84	5 to 50
B	2497.73	0.10 to 5
Ba	4554.04	10 to 300
Be	2348.61	0.1 to 3
Bi	3067.72	5 to 50
Ca	4226.73	1 to 50
Cd	2288.02	0.15 to 5
Co	3453.51	5 to 50
Cr	2843.25	10 to 100
Cu	3247.54	1 to 10
Fe	2462.64	10 to 300
In	3256.09	5 to 50
Mg	2779.83	10 to 100
Mn	2605.69	5 to 50
Mo	3132.59	0.5 to 10
Na	3302.32	80 to 400
Ni	3050.82	5 to 100
P	2553.28	50 to 500
Pb	2833.07	5 to 50
Sb	2598.05	1 to 50
Si	2435.16	10 to 200
Sn	3175.02	5 to 50
Ti	3361.26	1 to 100
V	3183.41	1 to 100
Zn	3345.02	20 to 300
Zr	3438.23	25 to 300

<sup>A</sup>All of the above lines are photographed in the second order, except barium and calcium which are first order lines.

<sup>B</sup>A gallium oxide carrier must be used for silver. See Test Method E 402.

with a spectrochemically pure carrier consisting of 16.4 mol % strontium fluoride in silver chloride. A given quantity of this mixture is placed in a special cupped electrode and excited in a d-c arc. The spectrum is recorded on photographic plates and the selected lines are either visually compared with standard plates or photometrically measured and compared with synthetically prepared standards exposed on the same plate.

### 39. Significance

39.1 Carrier distillation methods for the analysis of uranium over the past years have used a variety of carriers. Method E 402, approved by ASTM Committee E-2 on Emission Spectroscopy, called for gallium oxide as the carrier. This method involves the use of a mixture of silver chloride and strontium fluoride (17, 18). The fluoride gives an increased sensitivity for aluminum, zirconium, titanium, and niobium.

39.2 For the analysis of refractory elements in uranium, a separation is required for maximum sensitivity. However, recent work (19, 20) has improved the sensitivity of some elements using a mixed carrier technique.

### 40. Apparatus

40.1 *Spectrograph*—A spectrograph with sufficient resolving power and linear dispersion to separate the analytical lines from other lines in the spectrum of the sample in the spectral region 4200 to 7000 Å is required. Instruments with a reciprocal linear dispersion of approximately 5 Å/mm, first order or less, are satisfactory. A direct-reading spectrograph of comparable quality may be substituted for the equipment listed, in which case the directions given by the manufacturer

should be followed rather than those given in the succeeding steps of this procedure.

40.2 *Excitation Source*—Use a high-voltage spark source capable of providing a 14-A d-c arc (short circuit).

40.3 *Excitation Stand*—Conventional type with adjustable water-cooled electrode holders.

40.4 *Developing Equipment*—Use developing, fixing, washing, and drying equipment conforming to the requirements of Practice E 115 (21).

40.5 *Microphotometer*, having a precision of at least  $\pm 1\%$  for transmittances.

40.6 *Mixer*, for dry materials.<sup>15</sup>

40.7 *Platinum Crucible*, 10-mL capacity.

40.8 *Venting Tool*—See Fig. 3 for diagram.

40.9 *Calculating Boards*, or other special equipment are optional, their use depending to a large extent on how frequently analyses are made and how much speed is required.

40.10 *Muffle Furnace*, capable of heating up to 900°C.

<sup>15</sup> The Fisher-Kendall mixer was found to be satisfactory for large quantities and the Wig-L-Bug (Spex Industries) for small quantities.

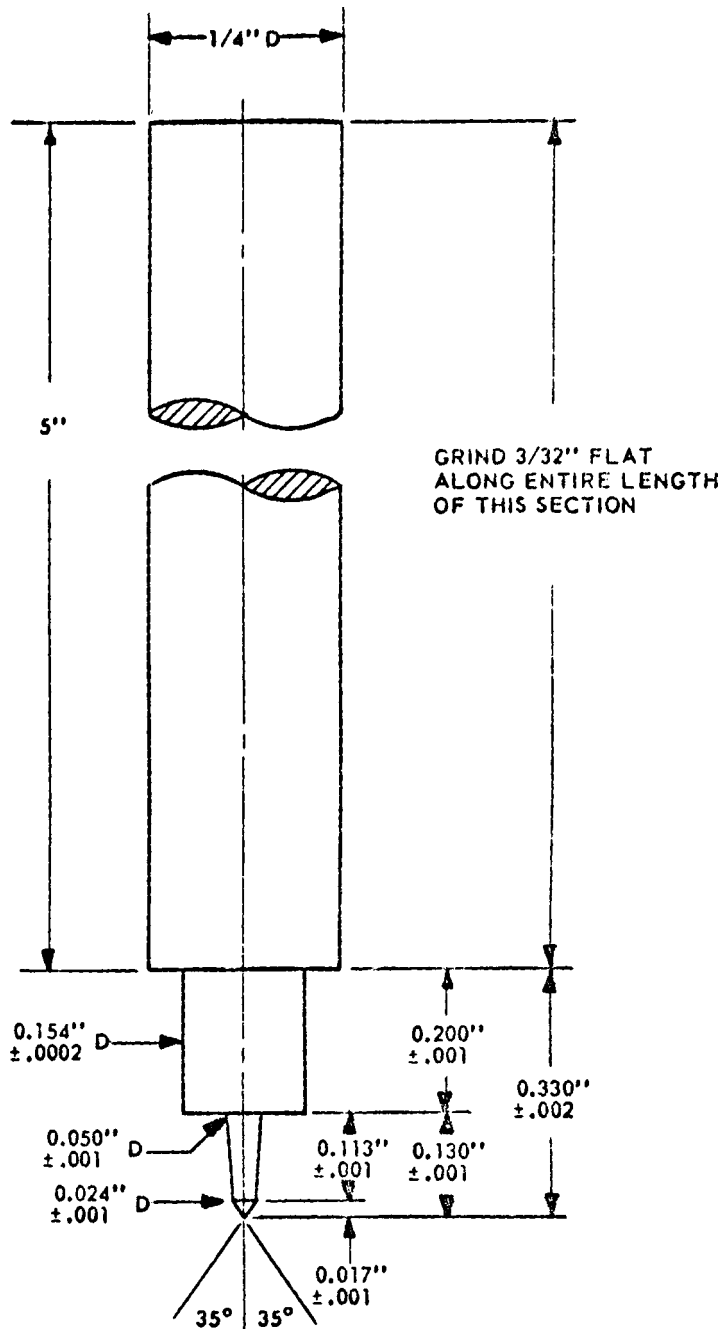


FIG. 3 Venting Tool



40.11 *Electrode Forceps*, with each V-tip bent to form a semicircular grasp around the electrodes.

40.12 *Balances*, torsion-type, one with a capacity up to 1 g and capable of weighing to  $\pm 0.1$  mg, and one with a capacity of 500 g.

## 41. Reagents and Materials

41.1 *Agate Mortars*.

41.2 *Electrodes*—The anode, pedestal, and counter electrodes should be respectively of the S-1, S-2, and C-1 types as given in Practice E 130.<sup>16</sup>

41.3 *Glassine Paper*.

41.4 *Tissue*—A suitable wiping tissue is necessary.

41.5 *Mixing Vial*, plastic, having a 12.7-mm (1/2-in.) diameter and a 25.4-mm (1-in.) length with cap, and a 9.6-mm (3/8-in.) diameter plastic ball.

41.6 *Nitric Acid* (HNO<sub>3</sub>, sp gr 1.42).

41.7 *Photographic Processing Solutions*—Prepare solutions as noted in Practice E 115.

41.8 *Silver Chloride-Strontium Fluoride Carrier* (16.4 mol % SrF<sub>2</sub> in AgCl<sup>17</sup>)—Since AgCl decomposes when exposed to light, all grinding, sieving, and transferring operations involving this material must be done in a darkroom under the safelight<sup>18</sup> and all blending must be done in opaque polyethylene bottles.

41.9 *Standard U<sub>3</sub>O<sub>8</sub> Diluent*—Use NBS SRM 950b U<sub>3</sub>O<sub>8</sub> or its replacement of known impurity content as a diluent.

41.10 *Photographic Film*—Use photo emulsion EK SA No. 1 or equivalent.

## 42. Standards

42.1 Standards can be synthesized by adding the impurity elements to purified U<sub>3</sub>O<sub>8</sub> (NBS SRM 950b) and homogenizing. Impurities in a solid or powder form, preferably as oxides, may be blended in U<sub>3</sub>O<sub>8</sub>, impurities in solution may be added to U<sub>3</sub>O<sub>8</sub> and the mixture dried, blended, and reignited, or the impurities and uranium may be combined in solution and reconverted to U<sub>3</sub>O<sub>8</sub>. The individual elements should grade in such a ratio as to facilitate visual comparisons, covering the desired analytical range for each. No single standard should have a total concentration of impurities exceeding 2000 µg/g. The bulk densities of the standards and the sample U<sub>3</sub>O<sub>8</sub> should be as nearly identical as possible.

42.2 The elements or compounds used to make U<sub>3</sub>O<sub>8</sub> impurity standards should be of the highest purity.

## 43. Procedures

43.1 *Preliminary Sample Preparation*:

43.1.1 Clean a 10-mL platinum crucible in HNO<sub>3</sub> (sp gr 1.42). Rinse with distilled water and dry. Transfer approximately 3 to 5 g of the UO<sub>2</sub> sample to a clean platinum crucible and heat in a muffle furnace at 800°C for 30 min. Remove from furnace and cool.

43.1.2 Grind the U<sub>3</sub>O<sub>8</sub> residue in an agate mortar and transfer to a clean labeled sample vial.

43.2 *Preparation of Electrode Charge*:

43.2.1 Weigh  $450 \pm 2$  mg of the sample as U<sub>3</sub>O<sub>8</sub> and transfer to a plastic mixing vial containing a plastic ball.

43.2.2 Perform operations at this point rapidly to minimize exposure to light. Cover the sample with a dark cover if possible. Weigh  $50 \pm 0.5$  mg of the silver chloride-strontium fluoride carrier and transfer to the same mixing vial.

43.2.3 Mix by rolling the vial between the fingers, and then process in the mixer for 30 s.

43.2.4 Weigh  $100 \pm 1.0$  mg of this mixture and transfer it into an S-2 graphite electrode. (Grade U-7 or equivalent).

43.2.5 Load duplicate electrodes for each sample and the plate standards. Use an electrode board to hold the electrodes, and identify the sample in each electrode by marking the board with the corresponding sample numbers.

43.2.6 To hold the electrodes use only clean forceps reserved for this purpose. Discard any electrodes accidentally touched or dropped.

43.2.7 Firmly grip the electrode with the modified forceps and pack the charge by gently tapping on a glassine-covered solid surface.

43.2.8 Further, compress and vent the charge with the venting tool shortly before arcing the sample. Wipe the venting tool point with a wiping tissue between different samples.

NOTE 9—**Caution:** Use extreme care to prevent jarring the electrodes after venting.

43.2.9 On a plate envelope, list the samples in the order in which they will be exposed and the spectrographic conditions.

43.3 *Exposure*:

43.3.1 Wipe the upper and lower electrode clamps with a wiping tissue before use. Place a pedestal and upper electrode in the appropriate clamps. Place the lower electrode firmly on the pedestal without jarring.

43.3.2 Expose the plate standards in order to obtain a line for the emulsion calibration curve.

43.3.3 Close the arc-enclosure door and critically adjust the electrodes to the 4-mm gap setting as indicated on the viewing screen.

43.3.3.1 *Exposure Conditions*:

Spectral range, Å	2250–5000
Slit width	optimum for the spectograph used
Preburn, s	0
Exposure, s	40
Current, A (short-circuit)	14
Voltage, V (open circuit)	250

43.3.4 Initiate the arc.

43.3.5 During the exposure continuously maintain the critical alignment of the arc image to the proper index lines on the viewing screen until the arc is automatically terminated.

43.3.6 Rack the plate holder for the next exposure. Drop spent electrodes into the container in the arc enclosure. Use a new upper electrode for each sample electrode arced. Replace the pedestal after 10 electrodes have been arced.

43.3.7 Repeat the exposure cycle until all the electrodes have been arced.

43.3.8 Rack the plate holder up to the end of travel and remove for processing.

<sup>16</sup> Upper electrode, Ultra Carbon 1992, lower electrode, Ultra Carbon 1998, electrode pedestal, Ultra Carbon 1993.

<sup>17</sup> Mallinckrodt A. R. AgCl and Spex Industries N. 1153 SrF<sub>2</sub>.

<sup>18</sup> The Eastman Safelight Filter, Wratten Series 1, has been found satisfactory.

#### 43.4 *Photographic Processing:*

43.4.1 Process the photographic plate in accordance with Practice E 115.

#### 43.5 *Photometry and Calculation of Results:*

43.5.1 With the microphotometer, measure the transmittance of the analytical lines and the adjacent background. Measure an appropriate step yielding between 15 and 75 % transmittance.

43.5.2 Measure the transmittance at seven steps of a suitable unfiltered line for the purpose of preparing an emulsion calibration curve. Repeat.

43.5.3 Plot the mean transmittance values on the *y*-axis versus the corresponding step numbers of the *x*-axis. Carefully draw a smooth curve through the points. Use linear graph paper.

43.5.4 Clip the emulsion calibration curve to the calculating board and determine the relative intensity, corrected for background, on the measured analytical lines for each standard and sample.

43.5.5 Obtain the results in  $\mu\text{g/g}$ ,  $\text{UO}_2$  basis, for each element in each sample from the appropriate analytical curve with reference to the plate standard.

#### 44. Precision and Accuracy

44.1 *Precision*—The relative standard deviation is 25 %.

44.2 *Accuracy*—The accuracy of the test method can approach the precision provided the appropriate standards are used.

### SILVER, SPECTROCHEMICAL DETERMINATION OF, BY GALLIUM OXIDE CARRIER D-C ARC TECHNIQUE

#### 45. Scope

45.1 This test method covers the spectrochemical determination of silver in nuclear-grade uranium dioxide. The relative standard deviation is 15 % for the concentration range of 0.1 to 50  $\mu\text{g/g}$ .

#### 46. Summary of Test Method

46.1 The uranium dioxide is ignited to  $\text{U}_3\text{O}_8$ , weighed, and mixed with gallium sesquioxide ( $\text{Ga}_2\text{O}_3$ ) in the ratio of 98 parts of  $\text{U}_3\text{O}_8$  to 2 parts of  $\text{Ga}_2\text{O}_3$ , and an appropriate internal standard is added to the mixture. The mixture is placed in a special cupped electrode and excited in a d-c arc. The  $\text{Ga}_2\text{O}_3$  carries silver (Ag), as a vapor or particulate, into the arc stream for excitation. The spectrum is recorded on a photographic plate and the selected silver lines are compared with standard plates of silver prepared according to standard spectrochemical procedures. Consult Test Method E 402 for procedural details.

### RARE EARTHS BY COPPER SPARK-SPECTROCHEMICAL METHOD

With appropriate sample preparation ICP-AES as described in C 761 or Inductively Coupled Plasma Mass Spectrometry (ICP-MS) as described in C 1287 may be used to determine rare earths and impurity elements.

### IMPURITY ELEMENTS BY A SPARK-SOURCE MASS SPECTROGRAPHIC METHOD

With appropriate sample preparation ICP-AES as described in C 761 or Inductively Coupled Plasma Mass Spectrometry (ICP-MS) as described in C 1287 may be used to determine rare earths and impurity elements.

### SURFACE AREA BY NITROGEN ABSORPTION METHOD

#### 47. Scope

47.1 This procedure is designed for a rapid determination of surface area of nuclear-grade uranium dioxide ( $\text{UO}_2$ ) powders. The determination of surface area by this procedure is automatic, simple, and fast enough to be used in quality control work. The range of analysis is from 1 to 1500  $\text{m}^2/\text{g}$ .

#### 48. Summary of Test Method

48.1 The surface area of  $\text{UO}_2$  powder is measured by low temperature gas adsorption using a surface area analyzer. The instrument is designed to give equilibrium adsorption at a predetermined relative pressure. Corrections are automatically made for sample bulb "dead space" and for the intercept on the ordinate of the multipoint Brunauer, Emmett, Teller (B.E.T.) plot. Nitrogen gas is used as the adsorbate. Automatic programming of the instrument produces a direct digital presentation of total surface area after equilibrium adsorption has occurred at a pre-set pressure and at liquid nitrogen temperature.

#### 49. Apparatus and Equipment

49.1 *Surface Area Analyzer*.<sup>19</sup>

49.2 *Nitrogen Gas Tank*, with a regulator, and pressure supplied to the instrument between 34 and 69 kPa gage (5 and 10 psig).

49.3 *Sample Bulb*, 15- $\text{cm}^3$  capacity.<sup>20</sup>

49.4 *Sample Tube Filler Funnel*.<sup>21</sup>

49.5 *Heating Mantle*, with thermocouple.<sup>22</sup>

49.6 *Dewar Flasks*, two, 500-mL (1-pt) volume.<sup>23</sup>

49.7 *Liquid Nitrogen*.

49.8 *Crushed Ice*.

49.9 *Drying Oven*.

49.10 *Vacuum Manifold*.<sup>24</sup>

49.11 *Mechanical-Vacuum Pump*.<sup>25</sup>

<sup>19</sup> A Micromeritics Surface Area Analyzer, Model 2200, has been found satisfactory.

<sup>20</sup> Micromeritics No. 04-61002 has been found satisfactory.

<sup>21</sup> Micromeritics No. 04-25846 has been found satisfactory.

<sup>22</sup> Micromeritics No. 03-26019 has been found satisfactory.

<sup>23</sup> Micromeritics No. 04-61001 has been found satisfactory.

<sup>24</sup> Numec AFA-409.

<sup>25</sup> Precision Scientific Co., Model 15, has been found satisfactory.

49.12 *Thermocouple Vacuum Gage*.<sup>26</sup>

## 50. Reagents and Chemicals

50.1 *Titanium Oxide* (TiO<sub>2</sub>) (10.3 m<sup>2</sup>/g).<sup>27</sup>

50.2 *Zinc Oxide* (ZnO), (3.9 m<sup>2</sup>/g).

## 51. Procedure

51.1 *Initial Setup:*

51.1.1 Turn master switch ON, CONTROL switch OFF, and SELECT switch to PREPARE.

51.1.2 Attach nitrogen to system.

NOTE 10—The nitrogen gas cylinder equipped with a pressure regulator capable of supplying 34 to 69 kPa gage (5 to 10 psig) pressure should be attached by means of heavy-walled or vacuum hose to the connector in the side of the instrument cabinet.

51.1.3 Set all three SAMPLE VALVES in PREPARE position for 5 min; then clockwise to OFF.

NOTE 11—All intermediate positions between labeled positions on sample valves are OFF positions.

(Master switch should have been on at least 10 min or more for instrument to operate properly in following steps):

51.1.4 Put the CONTROL switch in TEST.

51.1.5 Turn any one SAMPLE VALVE clockwise from OFF position to TEST. This will activate the counting mechanism.

51.1.6 Engage the RAPID-ADVANCE switch.

51.1.7 When the counting stops turn the SAMPLE VALVE clockwise to OFF between TEST and FILL.

51.1.8 Place the CONTROL switch in OFF position.

51.1.9 Place a Dewar flask of liquid nitrogen on sorption pump probe.

NOTE 12—Sorption pump probe is the short metal cylinder with the hemispherical end located immediately behind the sample-tube position.

51.1.10 Place the SELECT switch in TEST.

51.1.11 Leave for 5 to 8 min.

51.1.12 Place the SELECT switch in PREPARE.

51.1.13 Place the CONTROL switch to RESET position.

This will activate the motor which drives the piston upward.

51.1.14 Turn the SAMPLE VALVE clockwise to FILL and remove liquid nitrogen bath.

51.1.15 When piston stops upward travel or the red light on the instrument panel *just* goes out or both, turn the SAMPLE VALVE *immediately* clockwise to OFF. (If the red light goes out before the piston completes upward travel, turn the SAMPLE VALVE to OFF until piston stops, then turn the SAMPLE VALVE to FILL until red light *just* goes out again, then immediately to OFF.)

NOTE 13—Turning off immediately prevents gas from continuing to flow into the variable volume portion of the system and attaining a pressure too great for later operations.

51.1.16 Place the CONTROL switch to OFF position.

NOTE 14—The initial setup procedure is to fill the instrument with nitrogen gas. It needs to be repeated only when the master switch has been turned off or a fresh nitrogen cylinder attached to the instrument.

51.2 *Sample Preparation:*

51.2.1 Weigh an empty, clean, dry sample bulb.

51.2.2 Fill the sample bulb with approximately 6 g of sample and weigh.

NOTE 15—Sample masses must be such as to obtain 10 to 140 m<sup>2</sup> of surface. Best results are accomplished between 50 and 140 m<sup>2</sup> of surface. However, 30 m<sup>2</sup> of surface were measured for UO<sub>2</sub> without any loss of accuracy. This means that sample mass of UO<sub>2</sub> can vary between 5 and 10 g if the measured surface area is about 5 m<sup>2</sup>/g. For standard TiO<sub>2</sub>, use about 3 g. For standard ZnO, use about 6 g.

51.2.3 Outgas the sample under vacuum, at 200°C for 2 h using a separate vacuum manifold.<sup>28</sup>

NOTE 16—Degassing of UO<sub>2</sub> samples should be done on a separate manifold.

51.2.4 Remove the sample bulb from the manifold and attach the bulb to the instrument.

NOTE 17—Insert the sample tubes into the connectors in the recessed part of the instrument panel, being sure to push them all the way to the stop. Tighten the thumb nuts firmly by hand.

51.2.5 Turn the SAMPLE VALVE to PREPARE.

51.2.6 Place a heating mantle around the sample bulb and set the temperature to 150°C for 10 min.

NOTE 18—The sample has already been outgassed on a separate manifold but needs to be heated to release all gases adsorbed during the transfer of the sample. Heating is accomplished by purging nitrogen in order to drive off the released gases. Produce the gas flow by turning the SAMPLE VALVE to the PREPARE position. The outgassing temperature is indicated by the pyrometer at the upper left of instrument when the thermocouple probe of heating mantle is inserted into the jack labeled THERMOCOUPLE. Adjust the heating temperature by means of the variable transformer knob directly above the socket into which the mantle is plugged. (Setting of 35 to 40 will result in a temperature of about 150°C.)

51.2.7 Turn the SAMPLE VALVE clockwise to OFF between PREPARE and TEST.

51.2.8 Remove the heating mantle.

51.3 *Sample Analysis:*

51.3.1 Have the CONTROL switch in OFF,

51.3.2 Place the SELECT switch in PREPARE, and

51.3.3 Put the SAMPLE VALVE on OFF.

51.3.4 Place a Dewar flask of ice water around the sample bulb.

NOTE 19—The ice should be finely crushed and should be of sufficient quantity to encompass completely the sample bulb. Replenish the supply of ice, in the Dewar flask, two or three times a day during normal analysis conditions. The ice water level should be such that it just comes to the bottom of the frosted spot on the sample bulb. Stir the ice water before placing it on each sample and stir at least once during the time it is around the sample.

51.3.5 Turn the CONTROL switch to TEST.

51.3.6 Turn the SAMPLE VALVE to TEST position. The center red light should come on and the counter should indicate several counts. If this does not occur proceed to step 51.5.

51.3.7 Wait until the counter stops and the green light comes on.

<sup>26</sup> Bendix GTC-100 (range 0–1000 μm) has been found satisfactory.

<sup>27</sup> Material available from Particle Information Service, Los Altos, CA, has been found satisfactory.

<sup>28</sup> The Numec AFA-409 model Vacuum Manifold has been found satisfactory for this purpose.

51.3.8 Turn the SAMPLE VALVE counterclockwise to OFF position between PREPARE and TEST.

NOTE 20—The ice water bath procedure is used to establish a known quantity of gas in the sample bulb. That is why the preparation of ice-water slurry in the bath, the temperature, and the level of ice water are all very important.

51.3.9 Place the CONTROL switch to RESET until the motor stops, then to OFF.

51.3.10 Remove the ice water bath and dry the sample bulb.

51.3.11 Place the Dewar flask of liquid nitrogen around the sample tube making sure that the level of the liquid comes to the bottom of the frosted spot.

51.3.12 *Immediately* place SELECT switch and CONTROL switch in TEST positions.

51.3.13 In approximately 1 to 2 min, the red light will come on and the counter will run several counts.

51.3.14 After the counter stops, place the CONTROL switch in OFF position.

51.3.15 Zero the counter.

51.3.16 Place the CONTROL switch in TEST position.

51.3.17 Turn the SAMPLE VALVE to TEST position.

51.3.18 Engage RAPID ADVANCE switch.

NOTE 21—If there is a significant drop in the liquid nitrogen level while counting proceeds, add liquid nitrogen to maintain the level just at the bottom of the frosted spot. The Dewar flask should be covered with a styrofoam cup.

51.3.19 When equilibrium is reached, the green light will come on.

51.3.20 Record the reading on the counter as the total surface area of the sample in square metres.

51.4 *Reset Conditions:*

51.4.1 Turn the SAMPLE VALVE clockwise to OFF between TEST and FILL.

51.4.2 Set the CONTROL switch to OFF.

51.4.3 Turn the SELECT switch to PREPARE.

51.4.4 Set the CONTROL switch to RESET.

51.4.5 Turn the SAMPLE VALVE to FILL.

51.4.6 When the motor stops or the red light just goes out, or both, turn SAMPLE VALVE clockwise to OFF.

51.4.7 Remove liquid nitrogen.

51.4.8 Turn the SAMPLE VALVE clockwise to PREPARE.

51.4.9 Turn the CONTROL switch to OFF.

51.4.10 Allow the sample bulb to warm. When at room temperature turn the SAMPLE VALVE clockwise to OFF. Remove the sample bulb.

51.4.11 Weigh the sample bulb and sample. Subtract the mass of the sample bulb to obtain the mass of sample.

NOTE 22—The three SAMPLE VALVES have the same function. Thus, while one sample is being analyzed prepare two others for analysis, one of them being outgassed with heating mantle and the other having ice water bath around bulb.

51.5 *Correction for Overfill*—If in step 51.3.6, the red light did not come on and the counter did not indicate positive counts, the chamber has been overfilled with gas. To correct this condition proceed as follows:

51.5.1 Turn the SAMPLE VALVE clockwise to OFF position.

51.5.2 Set the CONTROL switch to OFF.

51.5.3 Place the liquid nitrogen bath on the sorption pump probe of a previously run sample.

51.5.4 Turn the SELECT switch to TEST position for 10 to 20 s.

51.5.5 Set the SELECT switch to PREPARE. The red light should come on. If it does not, repeat step 51.5.4.

51.5.6 Zero the counter.

51.5.7 Set the CONTROL switch to TEST.

51.5.8 Engage the RAPID ADVANCE switch.

51.5.9 Let counter run for 100 counts (10.0 m<sup>2</sup>).

51.5.10 Then turn the CONTROL switch to OFF.

51.5.11 If the red light is not off, turn the sample valve to FILL position until the red light goes off.

51.5.12 Turn the SAMPLE VALVE counterclockwise to TEST position.

51.5.13 Set the CONTROL switch to RESET until the red light comes on.

51.5.14 Turn the CONTROL switch back to TEST position.

51.5.15 Remove the liquid nitrogen.

51.5.16 Return to step 51.3.6 and proceed with the analysis.

## 52. Calculation

52.1 Divide the square metres obtained from step 51.3.20 of the procedure by the mass of the sample.

*Example:*

Number of square metres: 16.2

Mass of sample in g: 3.39

Surface area in m<sup>2</sup>/g: = 16.2/3.39 = 10.4

## 53. Precision and Accuracy

53.1 *Precision* is better than  $\pm 0.3$  m<sup>2</sup>/g.

53.2 *Accuracy* is within 2%.

### TOTAL GAS IN REACTOR-GRADE URANIUM DIOXIDE PELLETS

**This method was discontinued in January 1999.**

### THORIUM AND RARE EARTH ELEMENTS BY SPECTROSCOPY

**With appropriate sample preparation ICP-AES as described in C 761 or Inductively Coupled Plasma Mass Spectrometry (ICP-MS) as described in C 1287 may be used to determine rare earths and impurity elements.**

### HYDROGEN BY INERT GAS FUSION

## 54. Scope

54.1 This test method covers the determination of hydrogen over the range from 0.05 to 100  $\mu$ g of hydrogen (H<sub>2</sub>) in uranium dioxide UO<sub>2</sub> pellets by inert gas fusion.

## 55. Summary of Test Method

55.1 A UO<sub>2</sub> pellet heated in a graphite crucible in an inert-gas atmosphere to temperatures greater than 1800°C releases hydrogen, nitrogen, and oxygen (as carbon dioxide gas). After the interfering carbon dioxide and nitrogen are removed, the hydrogen is measured by integrating the output peak from a thermal-conductivity cell detector.

55.2 The instrument may be calibrated by either high-purity hydrogen or NBS SRMs.

## 56. Apparatus

56.1 *Automatic Hydrogen Determinator*,<sup>29</sup> consisting of an electrode furnace suitable for operation at 2400 to 2500°C; a carbon dioxide, oxygen, and nitrogen collection trap; a thermal conductivity cell for measuring hydrogen, and auxiliary purification systems.

56.2 *Crucibles*—Expendable graphite, 13 mm in outside diameter by 37 mm long.

56.3 *Argon Regulator*.

56.4 *Hydrogen Regulator*.

## 57. Reagents and Materials

57.1 *Argon (Ar)* (Industrial Grade, 99.999 %).

57.2 *Glass Wool*.

57.3 *High-Vacuum Silicone Lubricant*.

57.4 *Hydrogen (H<sub>2</sub>)* (Industrial Grade, 99.999 %).

57.5 *Schutzte Reagent* (iodine pentoxide over silica gel).

57.6 *Magnesium Perchlorate* (Mg(ClO<sub>4</sub>)<sub>2</sub>).

57.7 *Rare Earth Copper Oxide*.

57.8 *Sodium Hydroxide Over Asbestos*.

57.9 *Tin Flux*.

57.10 *Unalloyed Titanium Standard*, National Bureau of Standards SRM 352, 353, or 354.

## 58. Description of Instrument

58.1 The hydrogen determinator consists of three parts: (1) an electrode furnace for heating the sample, (2) a chemical and gas chromatographic system for separating the release gases, and (3) a thermal conductivity cell for measuring the hydrogen. Each part is discussed separately followed by a description of the gas system required for the operation of the instrument.

58.2 *Electrode Furnace*—The heating element of the electrode furnace consists of a graphite crucible (13 by 37 mm) held between two water-cooled copper electrodes. The graphite crucible is positioned by placing it on the lower electrode and the furnace is closed. The empty crucible is first outgassed at a temperature above 1800°C. The sample is then introduced into the crucible through a loading device located on the top of the furnace. The loading head consists of a plunger which, when pulled to the left, allows for sample introduction. The plunger is then pushed to the right and the sample is flushed with argon at room temperature. By rotating the plunger 180 deg, the sample drops into the crucible. When energized, a low-voltage, high-amperage current passes through the crucible, reaching a high temperature in a very short time. Under the thermal shock, the UO<sub>2</sub> pellet usually shatters into fragments 3 to 4 mm in diameter.

NOTE 23—The conditions listed in 57.2 may change with pellet size.

58.3 *Separation of Gases*—The gases released from a sample are primarily hydrogen, oxygen, and nitrogen. Water from the UO<sub>2</sub> pellet dissociates into hydrogen and oxygen at the high temperature in the presence of carbon. Oxygen and nitrogen interfere and are separated from the hydrogen before

it enters the thermal conductivity cell for measurement. Oxygen is converted to CO with the carbon from the graphite crucible. The CO is oxidized to CO<sub>2</sub> by means of a column filled with approximately 25 g of Schutze reagent (iodine pentoxide over silica gel). The CO<sub>2</sub> is then trapped in sodium hydroxide over asbestos. A magnesium perchlorate trap removes the water formed in the conversion of CO to CO<sub>2</sub>. Before entering the analytical train, the gases pass over a dust filter (glass wool) for removal of particulates. Nitrogen is separated by passing the gas through 3050-mm-long, 6.4-mm-diameter-wide molecular sieve 5A chromatographic column. Hydrogen gas is eluted first, followed by nitrogen gas with a 2-min separation time between the two gases.

NOTE 24—Depletion of the Schutze reagent is indicated by a change in color from yellow to dark brown and depletion of sodium hydroxide over asbestos is indicated by formation of a wet cake.

58.4 *Measurement of Hydrogen Gas*—The hydrogen gas eluting from the column enters the thermal conductivity cell where it is measured with reference to argon carrier gas. The large thermal conductivity difference between hydrogen and argon (ten times higher for hydrogen) is ideal for measurement of small amounts of hydrogen. The thermal conductivity cell consists of a pair of matched thermistors which constitute two legs of a Wheatstone bridge. The bridge becomes unbalanced and produces a signal which is integrated in the electronic system. The result of this integrated hydrogen peak is displayed as a positive reading on a digital voltmeter which is calibrated directly in micrograms of hydrogen per gram of UO<sub>2</sub> sample.

58.5 *Gas Flow System*—The argon carrier gas is purified before entering the analyzer. It passes over hot rare earth copper oxide (450°C) where hydrogen, if present, is converted into water. The water is removed by anhydrous and any CO<sub>2</sub>, if present, is removed by sodium hydroxide over asbestos. The purified argon gas is used in three separate gas flow systems: One portion of the purified argon gas is used to flush atmospheric gases from the furnace and contaminant gases from the crucible as it is heated during the outgas cycle. A second portion passes over the reference thermistor in the thermal conductivity cell. A third portion passes through the heated sample where the argon carries all the gases released from the sample to the analytical train. Hydrogen gas which may be used for calibration of the instrument first passes over sodium hydroxide over asbestos and magnesium perchlorate for removal of CO<sub>2</sub> and water, respectively. This gas bypasses the furnace by going directly to the thermal conductivity cell for measurement.

## 59. Operation of Instrument

59.1 After calibration and blank determination, the operation of the instrument involves loading an empty crucible in the furnace, dropping a sample in the loading device and dialing its weight on the weight compensator knobs. The operation then involves outgassing the empty crucible, by heating to above 1800°C for 45 s. The released gases are then expelled through the purge line for a duration of 25 s. The total outgas time is 70 s. After the OUTGAS cycle is completed, the sample is dropped into the outgassed crucible by rotating the handle on

<sup>29</sup> A commercially available unit, fully automatic and supplied with reagents, accessory parts, and standards are manufactured and sold by Laboratory Equipment Corp., St. Joseph, MI.

the loading device. The ANALYZE cycle begins with the sample being heated to greater than 1800°C for 120 s. (The UO<sub>2</sub> pellet usually shatters in the first 2 or 3 s.) Hydrogen is separated from the released gases and passes through the thermal conductivity cell, causing an imbalance in the bridge which results in a signal. The signal integration time is 120 s. At the end of the ANALYZE cycle, the hydrogen content of the sample is read directly in micrograms per gram on the digital voltmeter. The furnace is opened, the crucible containing the sample is removed and, after cleaning the furnace of carbon deposits on the upper and lower electrode, the instrument is ready for another sample.

### 60. Calibration with Metal Standards

60.1 Weigh an appropriate amount of NBS 352, 353, or 354 unalloyed titanium standard. Wash in acetone and dry before weighing.

60.2 Load and analyze the standards and record the results. Adjust the calibration controls in such a way as to produce the correct readout value on the direct readout meter. Analyze additional standards as needed to maintain the correct direct readout.

### 61. Startup Procedure

61.1 Follow the operating instructions provided by the manufacturer of the specific equipment used. After having properly set the operating controls of the instrument system, condition the apparatus by combusting several blanks using sample crucibles. Successive blank values should approach a constant value, allowing for normal statistical fluctuations.

NOTE 25—Determine a hydrogen blank on the tin flux by using 1 g of tin during blanking and calibration operations.

### 62. Procedure

62.1 Weigh a UO<sub>2</sub> pellet to the nearest 1 mg.

62.2 Place the UO<sub>2</sub> pellet into the loading chamber.

62.3 Drop the pellet into the furnace crucible and analyze as recommended by the instruction manual. Record the hydrogen value indicated by the direct readout meter.

### 63. Precision and Accuracy

63.1 The within-laboratory relative standard deviation of a single determination is 13 % within the range of UO<sub>2</sub> pellets having a hydrogen level from 0.13 to 0.20 µg. A bias cannot be determined since no hydrogen-certified UO<sub>2</sub> standards exist. The above information is provided for guidance as to the possible precision and accuracy of the analytical method, but it is not a guarantee of performance. The laboratory precision and accuracy of the method can only be determined by a good-in-laboratory measurement control program.

## URANIUM ISOTOPIC ANALYSIS BY MASS SPECTROMETRY

**This Test Method was discontinued in January 1999 and replaced by Test Method C 1413**

### 64. Keywords

64.1 impurity content; isotopic composition; stoichiometry; uranium content; uranium dioxide

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