



Standard Test Methods for Analysis of Sintered Gadolinium Oxide-Uranium Dioxide Pellets¹

This standard is issued under the fixed designation C 968; 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 analysis of sintered gadolinium oxide-uranium dioxide pellets to determine compliance with specifications.

1.2 The analytical procedures appear in the following order:

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1.3 The values stated in SI units are to be regarded as the standard.

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 696 Test Methods for Chemical, Mass Spectrometric, and Spectrochemical Analysis of Nuclear-Grade Uranium Dioxide Powders and Pellets³
- C 889 Test Methods for Chemical and Mass Spectrographic

Analysis of Nuclear-Grade Gadolinium Oxide (Gd₂O₃) Powder³

- C 922 Specification for Sintered Gadolinium Oxide-Uranium Dioxide Pellets³
- C 1347 Practice for Preparation and Dissolution of Uranium Materials for Analysis³
- C 1408 Test Method for Carbon (Total) in Uranium Oxide Powders and Pellets By Direct Combustion-Infrared Detection Method³
- C 1413 Test Method for Isotopic Analysis Of Hydrolysed Uranium Hexafluoride And Uranyl Nitrate Solutions By Thermal Ionization Mass Spectrometry³
- D 1193 Specification for Reagent Water⁴
- E 115 Practice for Photographic Processing in Optical Emission Spectrographic Analysis⁵
- E 116 Practice for Photographic Photometry in Spectrochemical Analysis⁵
- E 130 Practice for Designation of Shapes and Sizes of Graphite Electrodes⁵
- E 146 Methods for Chemical Analysis of Zirconium and Zirconium Alloys⁵

3. Significance and Use

3.1 The test methods in this method are designed to show whether a given material is in accordance with Specification C 922.

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 Reagent of the American Chemical Society, where such specifications are available.⁶ 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.

¹ These test methods are under the jurisdiction of ASTM Committee C-26 on Nuclear Fuel Cycle and are the complete responsibility of Subcommittee C26.05 on Test Methods.

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² Discontinued 1999. See C 968–94

³ *Annual Book of ASTM Standards*, Vol 12.01.

⁴ *Annual Book of ASTM Standards*, Vol 11.01.

⁵ *Annual Book of ASTM Standards*, Vol 03.05.

⁶ *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 Pharmacopoeia and National Formulary*, U.S. Pharmaceutical Convention, Inc. (USPC), Rockville, MD.

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

5. Safety Precautions

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

**CARBON (TOTAL) BY DIRECT COMBUSTION—
THERMAL CONDUCTIVITY METHOD**

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

**CHLORINE AND FLUORINE BY PYROHYDROLYSIS
ION-SELECTIVE ELECTRODE METHOD**

6. Scope

6.1 This test method describes the determination of chlorine and fluorine in gadolinium oxide-uranium dioxide pellets (Gd_2O_3/UO_2). With a 1 to 10-g sample, concentrations from 5 to 200 μg of chlorine and 1 to 200 μg of fluorine are determined without interference.

7. Summary of Test Method

7.1 The halogens are separated from the gadolinium oxide-uranium dioxide pellets by pyrohydrolysis in a quartz tube with a stream of wet oxygen sparge gas at a temperature of 900 to 1000°C (1, 2, 3, 4).⁷ Chlorine and fluorine are volatilized simultaneously as acids, absorbed in a buffer solution, and measured with ion-selective electrodes (4, 5, 6). Chloride can also be determined by amperometric titration.

8. Apparatus

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

8.1.1 *Gas Flow Regulator and Flowmeter.*

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

8.1.3 *Combustion Tube Furnace,* having a bore of about 32 mm (1 1/4 in.), a length of about 305 mm (12 in.), and the capability of maintaining a temperature of 1000°C.

8.1.4 *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.

8.1.5 *Combustion Boat*—A ceramic, platinum, or quartz boat with a 10-mL capacity, 89 to 102 mm (3 1/2 to 4 in.) long, 12.7 mm (1/2 in.) wide, and 9.53 mm (3/8 in.) high.

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

8.2 *Ion-Selective Electrodes*—A chloride-ion-selective activity electrode⁸ and a fluoride-ion-selective activity electrode.⁹

8.3 *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.

8.4 *Magnetic Stirrer.*

8.5 *Beakers,* 50-mL, polyethylene.

9. Reagents and Materials

9.1 *Accelerator*— U_3O_8 (halogen-free) can be used, but a flux of sodium tungstate (Na_2WO_4) with tungsten trioxide (WO_3) may be advantageous. (See Test Method C 696.) Special preparation of the mixture is necessary, that is, dehydrate 165 g of Na_2WO_4 in a large platinum dish. Transfer the dried material to a mortar. Add 116 g of WO_3 and grind the mixture to ensure good mixing. Transfer the mixture into a platinum dish and heat with a burner for 2 h. Cool the melt, transfer the flux to a mortar, and grind it to a coarse powder. Store the flux in an airtight bottle. Mix about 8 g of flux with each sample to be pyrohydrolyzed.

9.2 *Buffer Solution* (0.001 N)—Dissolve 0.1 g of potassium acetate ($KC_2H_3O_2$) in water, add 0.050 mL of acetic acid (CH_3CO_2H , sp gr 1.05), and dilute to 1 L.

9.3 *Chloride, Reference Solution* (1 mL = 100 μg Cl)—Dissolve 165 mg of dry sodium chloride ($NaCl$) in water and dilute to 1 L.

9.4 *Distilled Water*—Use ASTM Type IV water as specified in Specification D 1193.

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

⁷ The boldface numbers in parentheses refer to the list of references at the end of these methods.

⁸ The Orion Model No. 96-17 has been found satisfactory.

⁹ The Orion Method No. 9409 has been found satisfactory.

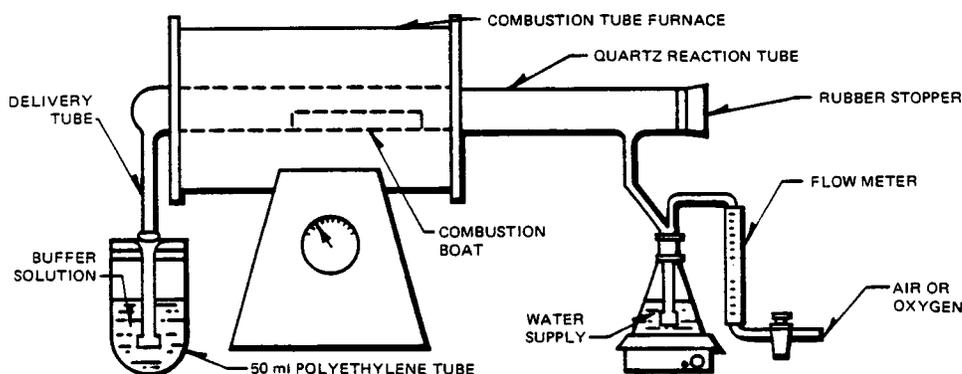


FIG. 1 Pyrohydrolysis of Gadolinium Oxide

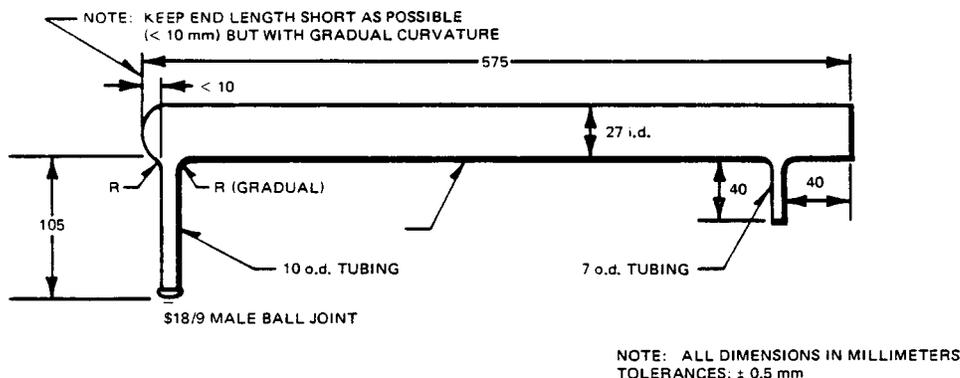


FIG. 2 Quartz Reaction Tube

9.6 Compressed Oxygen, Nitrogen, Helium, or Air.

10. Procedure

10.1 Adjust the pyrohydrolysis system to operating condition as follows:

- 10.1.1 Heat the furnace to 950 ± 50°C.
- 10.1.2 Fill the water reservoir and heat to 50 to 80°C.
- 10.1.3 Adjust the oxygen flow to about 1.5 to 2 L/min.

10.2 Flush the reaction tube and boat with moist oxygen in accordance with pyrohydrolysis procedures in 10.4.

10.3 Run a pyrohydrolysis blank using a halide-free uranium oxide or tungstate flux in accordance with the procedure in 10.4. A blank run should be made each day and after any sample that contains abnormally high levels of chloride or fluoride.

10.4 Sample Pyrohydrolysis:

10.4.1 Weigh 1 to 5 g of the crushed gadolinium oxide-uranium dioxide pellet and spread in the combustion boat. If an accelerator is desired, mix 4 g of U₃O₈ or 8 g of the tungstate flux with the Gd₂O₃/UO₂ before spreading in the boat.

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

10.4.3 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.

10.4.4 Check the oxygen flow and adjust to 1.5 to 2 L/min.

10.4.5 Continue the reaction for 1 h. (Thirty minutes may be sufficient with the tungstate flux.)

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

10.4.7 When the pyrohydrolysis is completed, transfer the buffer solution 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.

10.5 Determination of Chlorine and Fluorine with Ion-Selective Electrodes:

10.5.1 Assemble the meter and electrode in accordance with the instructions provided with the ion-selective electrode and the expanded scale meter being used.

10.5.2 Use successive dilutions of the chloride and fluoride reference solutions 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 10-mL aliquots of three or more references versus the concentration in micrograms per 25 mL on semi-log paper. The concentration of chloride should cover 10 µg/25 mL to 100 µg/25 mL and the fluoride from 5 µg/25 mL to 100 µg/25 mL.

11. Calculation

11.1 Calculate the chlorine and fluorine content as follows:

$$\text{Chlorine or fluorine, } \mu\text{g/g of Gd}_2\text{O}_3/\text{UO}_2 = (H_S - H_B)/W \quad (1)$$

where:

- H_S = halide in buffer solution + blank, µg,
- H_B = halide in pyrohydrolysis blank, µg, and
- W = sample mass, grams of Gd₂O₃/UO₂.

12. Precision and Bias

12.1 The relative standard deviation for the measurement of chlorine is 5 % in the range from 5 to 50 µg/g Gd₂O₃/UO₂ and increases to 10 % below the 5-µg/g level.

12.2 The relative standard deviation for the measurement of fluorine is 7 % in the range from 5 to 50 µg/g of Gd₂O₃/UO₂ and increases to 10 % for the range from 1 to 5 µg/g.

12.3 Recoveries from prepared reference spiked powder samples do not indicate the presence of a bias.

GADOLINIA CONTENT BY ENERGY-DISPERSIVE X-RAY SPECTROMETRY

13. Scope

13.1 This test method describes the determination of gadolinia in gadolinium oxide-uranium dioxide pellets (Gd₂O₃/UO₂) by energy-dispersive X-ray spectrometry. Concentrations from 0.4 to 10.0 % Gd₂O₃ may be determined with a 10-g sample pellet.

14. Summary of Test Method

14.1 X-ray fluorescence is a standard analytical technique in which an intense source of high-energy X rays excites atoms in the sample causing them to fluoresce or emit their characteristic X rays. The intensity of the emitted X rays is measured with a liquid-nitrogen-cooled, solid-state detector. The method is calibrated by comparing the measured intensity with that produced by reference materials of known gadolinia concentration at an averaged K α peak of 42.76 keV (7, 8).

14.2 This determination is carried out on an energy-dispersive X-ray spectrometer, where the radiation from an americium-241 or other source is used to activate the secondary radiation due to gadolinium and uranium. These secondary radiations can be detected with Si(Li), Ge(Li), or an intrinsic germanium solid state detector maintained at liquid nitrogen temperatures. By means of a single-channel analyzer the radiation is selected so as to record only those radiations due to the $K\alpha$ radiation of gadolinium.

14.3 This analysis may also be performed by wavelength dispersive X-ray analysis. The user must demonstrate the equivalency to the energy dispersive method.

15. Interferences

15.1 Rare earths interfere when concentrations are in excess of 1 % of Gd_2O_3/UO_2 .

16. Apparatus

16.1 *Solid-State X-Ray Detector*, $\geq 30\text{ mm}^2$ in area, $\geq 5\text{ mm}$ in thickness, with a resolution of 0.2 keV at 45 keV.

16.2 *Energy-Dispersive X-Ray Spectrometer System*—See Fig. 3.¹⁰

16.3 *Cryogenic Subsystem*, 17 L.¹⁰

16.4 *Annular X-Ray Source*—Americium-241 $\geq 0.10\text{ Ci}$.

16.5 *Collimator*—5-mm, machinable tungsten with 5 % copper.¹¹

16.6 *Diamond Blade Cut-Off Machine*, blade thickness 1.02 mm (0.040 in.).

16.7 *Grinding Wheel*, horizontally mounted.

16.8 *Ultrasonic Cleaner Apparatus*.

16.9 *Aluminum Sample Holder*—Inside diameter of the holder shall be of sufficient size to contain a plastic mounting ring 25.4 mm (1 in.) in diameter. This fit should be snug to align the sample geometrically and mount in the same position with each repetitive analysis.

17. Calibration Reference Materials

17.1 Pellet reference materials covering the weight percent range of interest must be carefully prepared. X-ray fluores-

cence excites only the surface atoms; hence differences in gadolinium content within each pellet must be no greater than 1 %. Gadolinia is hygroscopic and must be heated to assure $\leq 1\%$ water retention by weight when reference materials are initially prepared by blending weighed amounts of Gd_2O_3 and UO_2 .

17.2 The gadolinia content of the sintered pellet reference materials should be independently verified by another analytical method such as by oxalate precipitation (9).

18. Reagents and Materials

18.1 *Liquid Nitrogen*.

18.2 *Plastic Rings*, 25.4 mm (1-in.) diameter.

18.3 *Cap Plugs*, 25.4 mm (1-in.) diameter.

18.4 *Isopropyl Alcohol*.

18.5 *Epoxy Resin and Hardener*.

18.6 *Grinding Disks*, 320, 400, 600-grit.

18.7 *Gadolinium Oxide-Uranium Dioxide Reference Pellets*—Accurately prepare a series of working reference (Gd_2O_3/UO_2) sintered pellets covering the range of gadolinia concentrations anticipated in the pellets to be tested, using the purest gadolinia and uranium available.

19. Sample Preparation

19.1 It must be assured that samples and reference materials receive the same preparation. Sample holders and positioning devices will minimize geometrical effects when used properly.

19.2 Place sample or reference material of Gd_2O_3/UO_2 pellet longitudinally in the holding device of the diamond blade cut-off machine. Secure firmly but not so tight that mechanical damage will occur.

19.3 Cut the pellet in half using care not to force the cutting rate, as excessive pullout of the matrix will occur.

19.4 After sectioning, grind the pellet flat by hand using water as a coolant on 320-grit silicon carbide paper.

19.5 Clean ultrasonically in isopropyl alcohol; air dry.

19.6 Place polyethylene cap plug on the plastic ring forms.

19.7 Center the sectioned Gd_2O_3/UO_2 pellet in the plastic ring form.

19.8 Fill with proper mixture of epoxy and hardener and let stand till the mount sets.

19.9 After mounting operation, process the specimen using 320-grit grinding paper through the 500-grit series using water as a lubricant.

¹⁰ A commercially available unit, fully automatic, is manufactured and sold by KEVEX Corp., Burlingame, CA. Other systems may use component parts of other manufacturers.

¹¹ Material provided by Kulite Corp., Indianapolis, IN, has been found acceptable.

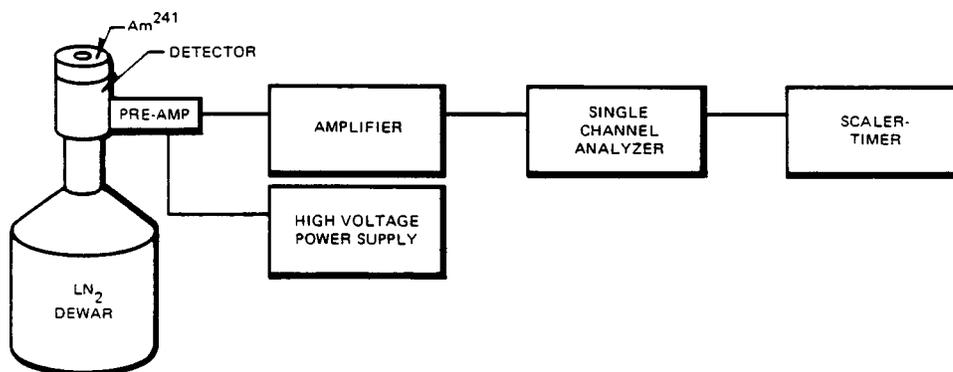


FIG. 3 Dispersive X-Ray Spectrometer System

19.10 Use medium pressure throughout the grinding operation with about 2 min spent on each grit size.

19.11 After final grinding, clean the sample ultrasonically in deionized water followed by isopropyl alcohol.

19.12 Dry with warm air.

20. Calibration

20.1 Place the mounted pellet standard specimen prepared in Section 28 into the aluminum sample holder. Place this on top of the americium-241 source of the energy-dispersive X-ray spectrometer.

20.2 Follow instructions manual for operation of instrument.

20.3 Set the instrument for a count of 10 000.

20.4 Record the reference material value and time in seconds for the 10 000 counts.

20.5 Calculate a least-squares straight line fit to the reference material data, converted to counts per second, versus concentration of gadolinia.

20.5.1 Calculate the “intercept” and “slope” constants of the pellet reference material as follows:

Intercept:

$$A = \frac{\sum C_i^2 \sum G_i - \sum C_i \sum C_i G_i}{N \sum C_i^2 - (\sum C_i)^2} \quad (2)$$

Slope:

$$B = \frac{N \sum C_i G_i - \sum C_i \sum G_i}{N \sum C_i^2 - (\sum C_i)^2} \quad (3)$$

where:

C_i = count rate of reference material i ,

= 10 000/ T_i , counts per second,

T_i = time for 10 000 counts, s,

G_i = weight percent of Gd₂O₃ in reference material i , and

N = number of reference materials.

21. Procedure for Unknown Samples

21.1 Follow 19.1 through 19.4 as required for Gd₂O₃/UO₂ reference material pellets. Always count a reference material with weight percent near nominal value of unknown.

21.2 Calculate the gadolinia weight percent using the calibration intercept and slope measured in 20.5.1.

21.3 Therefore:

$$\% \text{ Gd}_2\text{O}_3 = A + BC_i \quad (4)$$

22. Precision and Bias

22.1 The accuracy of the method is a function of the accuracy of the reference material and the care with which it is assured that no bias is produced between reference materials and samples. If the reference materials are accurate to <1 % and the bias between reference materials and samples is <1 %, then accuracy of about 1 % can be achieved.

22.2 The precision is a combination of the variances due to electronic drift, σ_e^2 ; counting variations, σ_c^2 ; geometry variations, σ_g^2 ; and calibration precision, σ_p^2 .

22.3 Therefore:

$$\text{Total } \sigma^2 = \sigma_e^2 + \sigma_c^2 + \sigma_g^2 + \sigma_p^2 \quad (5)$$

With care, standard deviations of 1 % are achievable for

each term; therefore, a total standard deviation of 2 % is possible.

HYDROGEN BY INERT GAS FUSION

23. Scope

23.1 This test method describes the determination of hydrogen in gadolinium oxide-uranium dioxide pellets (Gd₂O₃/UO₂) from all sources contained in the pellet. This measurement covers a range from 0.05 to 100 µg.

24. Summary of Test Method

24.1 A gadolinium oxide-uranium dioxide pellet is heated in a graphite crucible in an inert-gas atmosphere to temperatures greater than 1800°C. At that temperature, hydrogen, nitrogen, and oxygen (as carbon dioxide gas) are released. After the interfering carbon dioxide and nitrogen are removed, the hydrogen is measured by integrating the output peak from a thermal-conductivity cell detector.

24.2 The instrument may be calibrated using either high-purity hydrogen or NIST SRM 352C or equivalent material standards.

25. Apparatus

25.1 *Automatic Hydrogen Determination*,¹² consisting of an electrode furnace suitable for operation up to 2500°C; carbon dioxide, oxygen, and nitrogen collection traps; a thermal conductivity cell for measuring hydrogen, and an auxiliary purification system.

25.2 *Crucibles, Expendable Graphite*, 13 mm in outside diameter by 37 mm long.

25.3 *Argon Regulator*.

25.4 *Hydrogen Regulator*.

26. Reagents and Materials

26.1 *Argon* (Ar), industrial grade, 99.999 %.

26.2 *Glass Wool*.

26.3 *High-Vacuum Silicone Lubricant*.

26.4 *Hydrogen* (H₂), industrial grade, 99.999 %.

26.5 *Schutze Reagent*—Iodine pentoxide on silica gel.

26.6 *Magnesium Perchlorate* —[Mg(ClO₄)₂].

26.7 *Rare Earth Copper Oxide*.

26.8 *Sodium Hydroxide Over Asbestos*.

26.9 *Tin Flux*.

26.10 *Unalloyed Titanium Reference Material*—National Institute of Standards Technology SRM 352C, or equivalent material standards.

27. Operation of Instrument

27.1 Follow the operating instructions provided by the manufacturer of the specific equipment used. (See Test Methods C 696.) 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.

¹² A commercially available unit, fully automatic and supplied with reagents, accessory parts, and standards, is manufactured and sold by Laboratory Equipment Corp., St. Joseph, MI.

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

28. Calibration with Metal Reference Materials

28.1 Weigh an appropriate amount of NIST SRM 352C unalloyed titanium. Wash in acetone and dry before weighing. Other certified standards such as hydrogen in steel may be used.

28.2 Load and analyze the reference materials 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 reference materials as needed to maintain the correct direct readout.

29. Procedure

29.1 Weigh a whole Gd_2O_3/UO_2 pellet to the nearest 1 mg.

29.2 Place the pellet into the loading chamber.

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

30. Precision and Bias

30.1 The within-laboratory relative standard deviation of a single determination is 15 % for Gd_2O_3/UO_2 pellets having a hydrogen level from 0.1 to 0.25 $\mu\text{g/g}$. A bias cannot be determined since no hydrogen-certified Gd_2O_3/UO_2 reference materials 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 bias can only be determined by using surrogate hydrogen in metal standards in a measurement control program.

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

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

Samples can be dissolved using the appropriate dissolution techniques described in Practice C 1347

NITROGEN BY DISTILLATION—NESSLER REAGENT (PHOTOMETRIC) METHOD

31. Scope

31.1 This test method describes the determination of nitrogen in gadolinium oxide-uranium dioxide pellets (Gd_2O_3/UO_2). With a 2 to 5-g sample, concentrations from 5 to 100 μg of nitrogen are determined without interference.

32. Summary of Test Method

32.1 Pellet samples of gadolinium oxide-uranium dioxide are crushed, then dissolved in phosphoric acid. Hydrochloric acid with hydrogen peroxide can also be used. The resulting solution is made alkaline with sodium hydroxide, and the nitrogen is separated as ammonia by steam distillation (see Method E 146). Nessler reagent is added to the distillate to

form the yellow ammonium complex, and the absorbance of the solution is measured at approximately 430 nm, using a cell depth of 2 cm (14, 15).

NOTE 2—This procedure has been written for a cell having a 2-cm light path. The range of the method can be extended by suitably varying sample mass, aliquot size, amounts of reagents, and cell depth.

33. Interferences

33.1 There are no known interfering elements.

34. Apparatus

34.1 *Nitrogen Distillation Apparatus*, with 100-mL flask, Fig. 4; micro-Kjeldahl apparatus.

34.2 *Photometer*—A filter photometer with a narrow-band filter; or a *spectrophotometer* equipped with 2-cm cells.

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

35. Reagents and Materials

35.1 *Nessler Reagent*—Dissolve 50 g of potassium iodide (KI) in a minimum of cold water (approximately 35 mL). Add a saturated solution of mercuric chloride ($HgCl_2$) slowly until the first slight precipitate of red mercuric iodide persists. Add 400 mL of potassium or sodium hydroxide solution (505 g of KOH or 360 g of NaOH/L). Dilute the solution to 1 L with ammonia-free water, mix, and allow the solution to stand overnight. Decant the supernatant liquid and store it in a brown bottle. This reagent is stable indefinitely.

35.2 *Ammonium Chloride* (NH_4Cl)—Dry the ammonium chloride at 110 to 120°C for 2 h.

35.3 *Nitrogen Reference Solution* (1 mL = 10 mg N)—Dissolve 3.819 g of dried NH_4Cl in water and dilute the solution to 1 L. Transfer 10 mL of this solution to a 1-L volumetric flask and dilute it to volume with water.

35.4 *Hydrochloric Acid* (6 N)—Dilute 6 volumes of concentrated hydrochloric acid (HCl) to 11 volumes.

35.5 *Hydrogen Peroxide* (30 %).

36. Precautions

36.1 The use of ammonia or other volatile nitrogenous compounds in the vicinity of the experiment can lead to serious errors. To ensure freedom from contamination, take the following precautionary measures:

36.1.1 Steam clean all glassware immediately prior to use.

36.1.2 Use ammonia-free water in all cases.

37. Purity of Water

37.1 Unless otherwise indicated, all references to water in this method shall be understood to mean ammonia-free water, prepared as follows: Pass distilled water or other water of equivalent purity through a mixed-bed resin demineralizer. Prepare all solutions in an ammonia-free atmosphere and store them in tightly stoppered chemical-resistant glass bottles. Boil all rubber stoppers used for 30 min in sodium hydroxide solution (100 g NaOH/L) and rinse them with ammonia-free water.

38. Preparation of Calibration Curve

38.1 *Calibration Solutions*—Pipet 5, 10, 25, 50, 100, and 150 μg of the reference nitrogen solution (1 mL = 10 μg N)

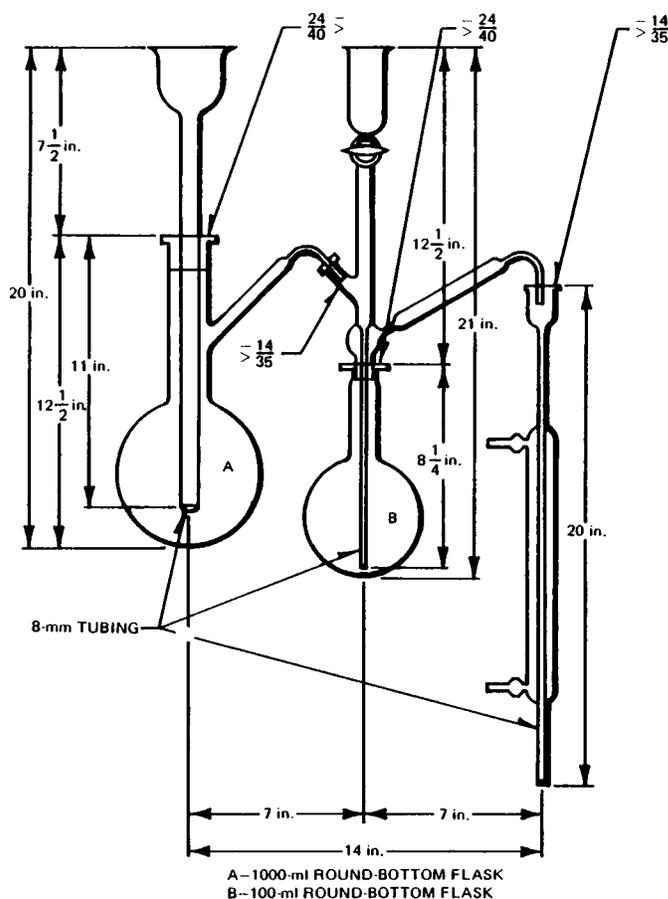


FIG. 4 Micro-Kjeldahl Apparatus

into 50-mL volumetric flasks containing 25 mL of water. Pipet 1.0 mL of Nessler reagent into each flask and dilute to the mark with water. Stopper the flask and mix well.

38.2 *Reference Solution*—Pipet 1.0 mL of Nessler reagent into a small volume of water in a 50-mL volumetric flask. Dilute to volume with water. Stopper and mix well.

38.3 *Photometry*—Transfer a suitable portion of water to a 2-cm absorption cell and adjust the photometer to the initial setting, using a light band centered at approximately 430 nm. While maintaining this photometer adjustment, take the photometric readings of the reference solution and the calibration solutions.

38.4 *Calibration Curve*—Correct for the blank (reference solution) reading and plot the photometric readings of the calibration solutions against micrograms of nitrogen per 50 mL of solution.

39. Procedure

39.1 Test Solution:

39.1.1 Weigh and transfer to a 500-mL Erlenmeyer flask 2 to 3 g of the crushed gadolinium oxide-uranium dioxide pellet sample. (The pellets should be crushed to a fine powder with a stainless steel mortar and pestle.)

39.1.2 Add to the distillation flask 60 mL of 6 N hydrochloric acid and 20 mL of 30 % hydrogen peroxide.

39.1.3 Boil for 10 min. While the solution is boiling, carefully add additional 30 % hydrogen peroxide dropwise

until the solids are dissolved and a clear yellow solution is obtained; cool the solution.

39.1.4 While the samples are dissolving, fill the steam-generating flask of the distillation unit with water. Apply heat and pass steam through the distillation flask and into the condenser. Collect 50-mL portions of the distillate and add 1.0 mL of Nessler reagent to each. When the absorbance of these solutions shows the apparatus to be free of ammonia, the distillation unit is ready for use with the samples.

39.1.5 Transfer quantitatively the dissolved gadolinia-urania solution into the 100-mL distillation flask.

39.1.6 Connect the distillation flask containing the sample solution to the distillation unit. Add 25 mL of NaOH solution (37.5 %) to the sample solution through the thistle tube so as to form two layers in the flask. Close the thistle tube stopcock and distill until 40 mL of distillate is collected. (The NaOH solution must be added slowly to avoid a violent reaction which may lead to a loss of sample.)

39.1.7 Pipet 1.0 mL of Nessler reagent to the distillate, dilute to the mark with water, stopper, and mix well.

39.2 *Reference Solution*—Carry a reagent blank through the entire procedure, using the same amount of all reagents.

39.3 *Photometry*—Take the photometric reading of the samples as described in 38.3.

40. Calculation

40.1 Correct for the blank and convert the photometric

reading of the sample to micrograms of nitrogen by means of the calibration curve.

40.2 Calculate the nitrogen content, N , in micrograms per gram of sample as follows:

$$N = A/W \quad (6)$$

where:

A = micrograms of nitrogen found, and

W = sample mass, grams of Gd_2O_3/UO_2 .

41. Precision and Bias

41.1 The relative standard deviation for the measurement of nitrogen at the 100 $\mu\text{g/g}$ level is 3 %.

OXYGEN-TO-METAL RATIO OF SINTERED GADOLINIUM OXIDE-URANIUM DIOXIDE PELLETS

42. Scope

42.1 This test method describes the determination of oxygen-to-metal ratio (O/M) of homogeneous sintered gadolinium oxide-uranium dioxide pellets (Gd_2O_3/UO_2), for small gadolinium concentrations.

43. Summary of Test Method

43.1 A gadolinium oxide-uranium dioxide pellet is crushed and preburned at 575°C for 2 h in air. It is then ignited at 900°C for 2 h in air, whereupon U-Gd-O phase composition close to $(U, Gd)_3O_8$ is attained. From the increase in weight due to oxygen pickup, the O/M of the pellets can be calculated (16, 17).

44. Interferences

44.1 A bias correction is required if localized high concentrations of Gd_2O_3 ($\geq 10\%$) in grains of $(U, Gd)O_2$ exist. This heterogeneity produces a lower weight change after ignition and a higher U^{+4} content that is observed with homogeneous gadolinia-uranium pellets, and must be corrected empirically (18).

45. Apparatus

45.1 *Muffle Furnace*, capable of maintaining a temperature of 575 \pm 25°C.

45.2 *Muffle Furnace*, capable of maintaining a temperature of 900 \pm 20°C.

45.3 *Indicating Pyrometers*, for above muffle furnaces.

45.4 *Analytical Balance*, with capability of 0.0001 g with measurement precision of ± 0.0001 g or better.

45.5 *Crucibles*, platinum, alumina, or porcelain, capacity 20 cm^3 .

45.6 *Desiccator*.

45.7 *Plattner's Mortar and Pestle*.

45.8 *Rotameters*, two, with valves capable of controlling at 500 cm^3 air/min.

45.9 *Pressure Pumps*, two, capable of delivering 500 cm^3 air/min.

46. Reagents and Materials

46.1 *Magnesium Perchlorate* — $[\text{Mg}(\text{ClO}_4)_2]$.

46.2 *Nitric Acid* (1 + 1), prepared by diluting 15.6 M HNO_3 with an equal volume of water.

47. Sample Preparation

47.1 Pellets must be crushed to a maximum of 3-mm (0.125-in.) diameter prior to analysis. Use Plattner's diamond mortar and pestle for crushing.

48. Operation of Furnace

48.1 Follow the operating instructions provided by the manufacturer of the specific furnace used.

49. Procedure

49.1 Weigh a crushed pellet into a clean, tared crucible that has been previously fired at 900°C for a minimum of 30 min.

49.2 Adjust the air flow to the 575 \pm 25°C muffle furnace to 500 cm^3/min .

49.3 Place the sample(s) on the hearth plate in the furnace and preburn at 575 \pm 25°C for 2 h at temperature.

49.4 After 2 h, transfer the sample(s) to the 900 \pm 20°C muffle furnace.

49.5 Adjust the air flow of the muffle furnace to deliver 500 cm^3/min of air.

49.6 Ignite the sample(s) for 2 h at temperature cool to room temperature.

49.7 Weigh the oxidized pellet sample(s).

49.8 Clean the crucibles in (1 + 1) nitric acid, rinse with deionized water and ignite at 900°C for subsequent analyses.

50. Calculation

50.1 Calculate the oxygen-to-metal ratio, O/M , as follows:

$$O/M = 2.667 - \left(\frac{W_2 - W_1}{W_2} \right) (17.54538 - 0.00190E + 0.01G) \quad (\text{Note 7}) \quad (7)$$

where:

W_1 = initial sample weight,

W_2 = sample weight after oxidation,

E = enrichment, %, and

G = Gd_2O_3 , %.

NOTE 3—Factors in this equation take into account the different levels of gadolinium and U-235 in Gd_2O_3/UO_2 fuel.

NOTE 4—Laboratories may need to develop equations that better fit their own product characteristics and accuracy requirements. This may include experimentally derived corrections for UO_2/Gd_2O_3 solid solutions at various concentrations, as well as impurities corrections.

51. Precision and Accuracy

51.1 Laboratory precision of accuracy cannot be determined since no oxygen-to-metal certified Gd_2O_3/UO_2 reference materials exist. The method can only be determined by a good in-laboratory measurement program on pellets from the same lot of material.

SPECTROCHEMICAL DETERMINATION OF TRACE, IMPURITY ELEMENTS

52. Scope

52.1 This test method describes the determination of the following trace element impurities in gadolinium oxide-uranium dioxide (Gd_2O_3/UO_2) pellets: aluminum, calcium,

chromium, cobalt, iron, magnesium, nickel, and silicon. The concentration range is given in Table 1.

53. Summary of Test Method

53.1 The as-received pellets are ignited to Gd_2O_3/U_3O_8 . The oxidized sample is mixed with a carrier consisting of gallium oxide, strontium fluoride, and indium oxide. The mixture is placed in an electrode and excited in a d-c arc. Varying amounts of impurities either in vapor form or as solid particles are carried up into the arc stream along with the vaporized gallium oxide mixture. The spectrum is recorded on a photographic plate and the selected lines are either visually compared with standard or reference plates, or photometrically measured and compared with synthetically prepared reference materials according to accepted spectrochemical procedures (19, 20, 21).

54. Apparatus

54.1 *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 from 225 to 450 nm. A reciprocal linear dispersion of 0.5 nm/mm, first order or less, is satisfactory. A direct-reading spectrometer 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.

54.2 *Excitation Source*, capable of providing a 14-A dc (short circuit).

54.3 *Excitation Stand*, conventional type, with adjustable water-cooled electrode holders.

54.4 *Developing Equipment*—Use developing, fixing, washing, and drying equipment conforming to the requirements of Practices E 115.

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

54.6 *Calculating Boards*, capable of converting percent transmission values into intensity or density values.

54.7 *Mixer Mill*, for dry materials.¹³

54.8 *Platinum Crucibles*, 25-mL capacity, or

54.9 *Coors Crucibles*, tall form, 30-mL capacity.

54.10 *Venting Tool*—See Fig. 5 for diagram.

54.11 *Muffle Furnace*, capable of heating to 1000°C.

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

54.13 *Balance*, torsion-type, with a capacity to 200 g and capable of weighing to ± 0.1 mg.

55. Reference Materials

55.1 Reference materials can be synthesized by adding the impurity elements to high purity U_3O_8 (NBL CRM 114 or equivalent) and homogenizing. Impurities in a solid or powder form, preferably as oxides, may be blended into the U_3O_8 . The individual elements should grade in such a ratio as to facilitate visual comparisons, covering the desired analytical range for each. No single reference material should have a total concentration of impurities exceeding 2000 $\mu\text{g/g}$. The bulk densities of the reference materials and the sample U_3O_8 should be as nearly identical as possible.

55.2 The elements of compounds used to make the U_3O_8 impurity reference materials should be of the highest purity.

56. Reagents and Materials

56.1 *Carrier*—Gallium oxide–strontium fluoride–indium oxide. Weigh into a large plastic vial 13 g of gallium oxide, 6 g of strontium fluoride, and 1 g of indium oxide. Add two plastic balls, seal with a cap, then blend the mixture on a mixer mill for 10 min.

56.2 *Electrodes*—The anode, pedestal, and the counter electrodes should be respectively of the S-1, S-2, and C-1 types as given in Practice E 130. The graphite should be a high-purity type with an average density near 1.85 g/cm^3 and a specific electrical resistance near $4.5 \times 10^{-4} \Omega\text{-in}$.

56.3 *Photographic Emulsion*, Eastman Kodak, Type SA No. 1, II-F and I-N plates or film or equivalent. Calibrate the emulsion in accordance with Practice E 116.

56.4 *Photographic Processing Solutions*—Formulas for processing solutions are given in Practices E 115.

56.5 *Mixing Vial*, plastic, having a 12.7-mm ($\frac{1}{2}$ -in.) diameter and a 25.4-mm (1 in.) length with a cap, and a 9.6-mm ($\frac{3}{8}$ -in.) diameter plastic ball.

56.6 *Mixing Vial*, plastic, having a 9.1-mm ($\frac{3}{4}$ -in.) diameter and a 51-mm (2-in.) length with cap, and 15-mm ($\frac{5}{8}$ -in.) diameter plastic ball.

56.7 *Standard U_3O_8 Diluent*—Use NBS CRM 114 U_3O_8 or equivalent of known impurity content as a diluent.

56.8 *Gadolinium Oxide*, high-purity (99.999 %).¹⁴

57. Procedure

57.1 *Preliminary Sample Preparation*:

57.1.1 Clean a 25-mL platinum crucible in HNO_3 (sp gr 1.42). Rinse with distilled water and dry. Transfer one Gd_2O_3/UO_2 pellet sample to a clean platinum crucible and heat in a muffle furnace at $400 \pm 25^\circ\text{C}$ for 1 h, then increase temperature to $900 \pm 25^\circ\text{C}$ for one additional hour. Remove from the furnace and cool.

57.1.2 Grind the Gd_2O_3/U_3O_8 mixture in an agate mortar and transfer to a clean labeled sample vial.

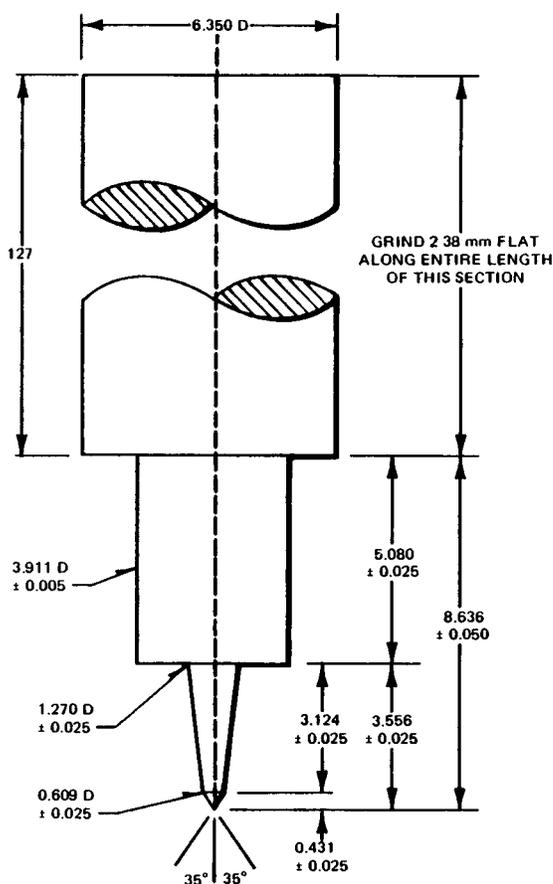
¹³ Mixer Mill, sold by Spex Industries, Inc., Metuchen, NJ, has been found satisfactory.

TABLE 1 Recommended Analytical Spectral Lines and Concentration Range of Trace Elements

Element	Analytical Line, nm	Concentration Range, $\mu\text{g/g}$ of U
Al	236 706	10–300
Ca	422 673 ^A	1–50
Cr	284 325	10–100
Co	345 351	5–50
Fe	246 264	10–300
Mg	277 983	10–100
Ni	305 082	5–100
Si	243 516	10–200

^ACalcium line must be photographed in the first order; all other lines are photographed in the second order.

¹⁴ Spex Industries, Inc., Metuchen, NJ, supplies high-purity gadolinium oxide that has met these requirements.



NOTE 1—All dimensions are in millimetres.

FIG. 5 Venting Tool

57.1.3 Weigh 1.000 ± 0.002 g of the Gd_2O_3/U_3O_8 sample and transfer to a plastic mixing vial containing a plastic ball.

57.1.4 Weigh 50 ± 0.5 mg of the gallium oxide-strontium fluoride-indium oxide carrier and transfer to the same mixing vial.

57.1.5 Mix by rolling the vial between the fingers, and then process in the mixer for 60 s.

57.1.6 Weigh 100 ± 1.0 mg of this mixture into a graphite electrode.

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

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

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

57.1.10 Further, compress and vent the charge with the venting tool point (Fig. 5), and clean the tips with a wiping tissue between different samples. Use extreme care to prevent jarring the electrodes after venting.

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

57.2 Exposure:

57.2.1 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.

57.2.2 Expose the plate reference materials in order to obtain a line for the emulsion calibration curve.

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

57.2.3.1 Exposure Conditions:

Spectral range, nm	225–450
Slit, width, μm	50
Preburn period, s	5
Exposure period, s	50
Current, A, d-c (short circuit)	14

57.2.4 Initiate the arc.

57.2.5 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.

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

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

57.2.8 Rack the plate holder up to the end of travel and

remove for processing.

57.3 *Photographic Processing*—Process the photographic plate in accordance with Practice E 115.

57.4 *Photometry and Calculation of Results*:

57.4.1 With the microphotometer, measure the transmittance of the analytical lines, as recommended in Table 1, and the adjacent background. Measure an appropriate step yielding between 15 and 75 % transmittance.

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

57.4.3 Plot the mean transmittance values on the *Y*-axis versus the corresponding steps numbers of the *X*-axis. Carefully draw a smooth curve through the points. Use linear graph paper.

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

57.4.5 Obtain the results in $\mu\text{g/g}$ of U basis, for each element in each sample from the appropriate analytical curve with reference to the plate reference materials.

57.4.6 Steps 57.4.2 through 57.4.4 are optional.

58. Precision and Bias

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

58.2 *Bias*—The bias of the method can approach the precision provided appropriate reference materials are used.

TOTAL GAS BY HOT VACUUM EXTRACTION This procedure was discontinued in January 1999.

CERAMOGRAPHIC DETERMINATION OF FREE Gd₂O₃ AND FREE UO₂ TO ESTIMATE THE HOMOGENEITY OF (U,Gd)O₂ PELLETS

59. Scope

59.1 The homogeneity of Gd₂O₃ in UO₂ has been cited in Specification C 922 as an important requirement for this fuel form. The uniform distribution of gadolinia in urania will result in up to three components in the pellet: free Gd₂O₃, free UO₂, and a Gd₂O₃-UO₂ solid solution. There are a number of ways for assessing uniformity of which the ceramographic method described here may not be the most definitive. This technique has been used over the gadolinia concentration range from 1 to 10 weight %.

60. Summary of Test Method

60.1 This ceramographic test, similar to those reported by Rooney (23) and by Hammerschmidt and Saiger (24), consists of making micrographs of polished pellet surfaces that have been chemically etched with a hydrogen peroxide-sulfuric acid solution. Oxidation makes it possible to differentiate between components.

61. Apparatus

61.1 *Diamond Saw*, low-speed.

61.2 *Ultrasonic Cleaner*.

61.3 *Polisher/Grinder Table*.

61.4 *Metallurgical Microscope* with camera attachment.

62. Reagents and Materials

62.1 *Hydrogen Peroxide* (30 %).

62.2 *Hydrogen Peroxide-Sulfuric Acid Etch Solution*—10 parts 30 % hydrogen peroxide mixed with 1 part concentrated sulfuric acid.

62.3 *Silicon Carbide Grinding Disks*, 240, 320, 400, and 600-grit.

62.4 *Diamond Paste*, 7, 3, and 1- μm grains.

62.5 *Color Film*.

63. Sample Preparation

63.1 Cut the fuel pellet in the longitudinal axis, somewhat off center, using a low-speed diamond saw.

63.2 Cast the larger pellet part in cold-setting resin.

63.3 Grind the cast specimen on a polisher/grinder table using successively, silicon carbide in four stages from 240 to 600 grit; then polish with diamond pastes in stages with 7, 3, and 1- μm grains.

63.4 Clean the specimen in an ultrasonic cleaner after each stage of grinding and polishing.

64. Sample Analysis

64.1 Generate a weak grain surface by applying a 1-min wiping etch to the polished surface with hydrogen peroxide-sulfuric acid solution.

64.2 Flush the sample with water, then alcohol, and dry with warm air.

64.3 Heat the specimen to 45°C and immediately dip into a 30 % hydrogen peroxide solution maintained at 45°C.

NOTE 5—Over-etched specimens must be reground and polished.

64.4 Allow the etch to develop for 3 min; 45 s. Touch the top of the peroxide solution every 30 to 40 s to dispel bubbles formed on the specimen surface.

64.5 Flush the sample with water, clean ultrasonically in alcohol, and dry with warm air.

64.6 Examine and photograph the samples under a metallurgical microscope at a magnification of 100 \times in white light (bright-field illumination with no filters).

65. Evaluation

65.1 Measure the weight percent of free gadolinia by scanning the entire micrograph surface for gadolinia inclusions (white-colored) and using the following calculation:

$$\text{Free Gd}_2\text{O}_3 \text{ wt } \% = \frac{A(0.72)100}{\% \text{ Gd}_2\text{O}_3 (A_c)} \quad (8)$$

where:

A = total observed surface of all Gd₂O₃ inclusions, cm²,
 A_c = total pellet surface examined without Gd₂O₃, cm³
 = nominal weight percent of Gd₂O₃, and
 0.72 = factor from the density ratio Gd₂O₃/(U,Gd)O₂.

65.2 Measure the weight percent of free uranium dioxide by means of a blue spot analysis on a representative color photo using a calibrated grid, counting the number of uranium particles covered by crosses and converting this count to

weight percent of free UO_2 . (Estimate the average particle size and assume sphericity to assign a particle weight.)

65.3 Calculate the weight percent of solid $(\text{U,Gd})\text{O}_2$ solution by difference:

$$\begin{aligned} w/o(\text{U,Gd})\text{O}_2 &= 100 - \text{wt\% free } \text{UO}_2 \\ &\quad - \text{wt\% free } \text{Gd}_2\text{O}_3 \left(\frac{\text{wt\% } \text{Gd}_2\text{O}_3}{100} \right) \end{aligned} \quad (9)$$

66. Precision and Bias

66.1 This test method is subjective, and inadequate data are available for a statistical evaluation.

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