Designation: C1000 – 11

Standard Test Method for
Radiochemical Determination of Uranium Isotopes in Soil by Alpha Spectrometry[1]

This standard is issued under the fixed designation C1000; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision. A number in parentheses indicates the year of last reapproval. A superscript epsilon (ε) indicates an editorial change since the last revision or reapproval.

1. Scope

1.1 This test method covers the determination of alpha-emitting uranium isotopes in soil. This test method describes one acceptable approach to the determination of uranium isotopes in soil.[2]

1.2 The test method is designed to analyze 10 g of soil; however, the sample size may be varied to 50 g depending on the activity level. This test method may not be able to completely dissolve all forms of uranium in the soil matrix. Studies have indicated that the use of hydrofluoric acid to dissolve soil has resulted in lower values than results using total dissolution by fusion.

1.3 The lower limit of detection is dependent on count time, sample size, detector, background, and tracer yield. The chemical yield averaged 78 % in a single laboratory evaluation, and 66 % in an interlaboratory collaborative study.

1.4 The values stated in SI units are to be regarded as standard. The values given in parentheses are for information only.

1.5 *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.* A specific precautionary statement is given in Section 10.

2. Referenced Documents

2.1 *ASTM Standards:*[3]

C859 Terminology Relating to Nuclear Materials
C998 Practice for Sampling Surface Soil for Radionuclides
C999 Practice for Soil Sample Preparation for the Determination of Radionuclides
C1163 Practice for Mounting Actinides for Alpha Spectrometry Using Neodymium Fluoride
C1284 Practice for Electrodeposition of the Actinides for Alpha Spectrometry
D1193 Specification for Reagent Water
D3084 Practice for Alpha-Particle Spectrometry of Water
D3648 Practices for the Measurement of Radioactivity
D7282 Practice for Set-up, Calibration, and Quality Control of Instruments Used for Radioactivity Measurements

3. Summary of Test Method

3.1 A soil sample with uranium-232 tracer added is heated to destroy organic matter and dissolved with a mixture of hydrofluoric acid and nitric acid. The uranium is coprecipitated with ferric hydroxide and the precipitate is dissolved with hydrochloric acid. Iron is removed by extraction with isopropyl ether, and plutonium, radium, and thorium are separated from uranium by anion exchange. Uranium is electrodeposited on a stainless steel disk and determined by alpha spectrometry. As an option, the uranium may be prepared for alpha spectrometric measurement by using coprecipitation with neodymium fluoride.

4. Significance and Use

4.1 This test method is used to analyze soil for alpha-emitting uranium isotopes. It can be used to establish baseline uranium levels and to monitor depositions from nuclear facilities.

5. Interferences

5.1 Protactinium-231 may not be completely separated by the procedure and could interfere with the determination of uranium-233 or uranium-234 because it has the following alpha energies in MeV: 5.06, 5.03, 5.01, 4.95 and 4.73 (see Appendix X1). If neptunium is present in the sample in the plus four oxidation state, it will co-elute with the uranium.

5.2 Since uranium-232 is added as a tracer, it can not be determined in soil. Uranium-232 is rarely present in soil samples. If present in significant quantities relative to the

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[1] This test method is under the jurisdiction of ASTM Committee C26 on Nuclear Fuel Cycle and is the direct responsibility of Subcommittee C26.05 on Methods of Test.

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[2] Casella, V. A., Bishop, C. T., and Glosby, A. A., "Radiometric Method for the Determination of Uranium in Soil and Air," U.S. Environmental Protection Agency, EPA-600/7-80-019, Las Vegas, NV, February 1980; and in Practices D3084 and D3648.

[3] For referenced ASTM standards, visit the ASTM website, www.astm.org, or contact ASTM Customer Service at service@astm.org. For *Annual Book of ASTM Standards* volume information, refer to the standard's Document Summary page on the ASTM website.

activity of uranium-232 tracer added, uranium-232 will lead to an overestimation of the chemical yield and a low bias in uranium results.

6. Apparatus

6.1 *Alpha Pulse Height Analysis System:*[2]

6.1.1 A system consisting of a charged particle detector capable of 50 keV or less resolution on samples electrodeposited on a flat mirror-finished stainless steel disk is required.

6.1.2 The resolution is defined as the width of an alpha peak when the counts on either side of the peak are equal to one-half of the counts at the maximum of the peak (full width at half maximum height (FWHM)).

6.1.3 The counting efficiency of the system should be greater than 15 % and the background in the energy region of each peak should be less than 0.010 cpm.

6.1.4 A regular program of measurement control operations should be conducted for the alpha spectrometry system such as regular background checks, daily source check to determine system stability, control charting, and careful handling of samples during changing. See Practice D7282.

6.2 *Beakers and Covers* (TFE-fluorocarbon), 250 mL.

6.3 *Porcelain Crucible,* 60 mL.

6.4 *Centrifuge and Bottles,* 250-mL capacity.

6.5 *Ion Exchange Columns,* 1.3 cm inside diameter by 15 cm long with 100 mL reservoir.

6.6 *Automatic Pipettes,* 0.1 to 1.0 mL with disposable tips.

6.7 *Furnace,* able to maintain 600°C.

7. Reagents

7.1 *Purity of Reagents*—Reagent grade chemicals shall be used in all tests. Unless otherwise indicated, it is intended that all reagents conform to the specifications of the Committee on Analytical Reagents of the American Chemical Society, where such specifications are available.[4] Other grades may be used, provided it is first ascertained that the reagent is of sufficiently high purity to permit its use without lessening the accuracy of the determination.

7.2 *Purity of Water*—Unless otherwise indicated, references to water shall be understood to mean reagent water as defined in Specification D1193, Type III.

7.3 Reagent purity shall be such that the measured radioactivity of blank samples does not exceed the calculated probable uncertainty of the measurement.

7.4 *Ammonium Hydroxide* (0.15 *M*)—Mix 10 mL of concentrated ammonium hydroxide with water and dilute to 1 L.

7.5 *Ammonium Hydroxide (sp gr 0.90)*—Concentrated ammonium hydroxide (NH_4OH). Keep tightly capped to minimize the uptake of carbon dioxide.

7.6 *Ammonium Sulfate Solution (1 M)*—Dissolve 132 g of $(NH_4)_2SO_4$ in water and dilute to 1 L.

7.7 *Anion Exchange Resin*[5]—Type 1 anion exchange resin, 8 % cross-linked, 100 to 200 mesh, chloride form. Prepare a resin slurry by soaking the resin in 8 M HCl and transfer the slurry to an ion exchange column so that the resin column is approximately 10 cm high.

7.8 *Ferric Chloride Solution* (0.18 *M* in 0.5 *M* HCl)—Dissolve 48 g of $FeCl_3$. $6H_2O$ in 0.5 *M* HCl and dilute to 1 L.

7.9 *Hydriodic Acid (48 %)*—Concentrated hydriodic acid (HI).

7.10 *Hydrochloric Acid* (0.5 *M*)—Mix 42 mL of concentrated HCl with water and dilute to 1 L.

7.11 *Hydrochloric Acid (1 M)*—Mix 83 mL of concentrated HCl with water and dilute to 1 L.

7.12 *Hydrochloric Acid (6 M)*—Mix 500 mL of concentrated HCl with water and dilute to 1 L.

7.13 *Hydrochloric Acid (8 M)*—Mix 667 mL of concentrated HCl with water and dilute to 1 L.

7.14 *Hydrochloric Acid (sp gr 1.19)*—Concentrated hydrochloric acid (HCl).

7.15 *Hydrochloric Acid-Hydriodic Acid Solution (HCl-HI)*—Mix 1 mL of concentrated HI with 50 mL of 6 *M* HCl. Prepare immediately before use.

7.16 *Hydrofluoric Acid (48 %)*—Concentrated hydrofluoric acid (HF).

7.17 *Nitric Acid (sp gr 1.42)*—Concentrated nitric acid (HNO_3).

7.18 *Uranium-232, Standard Solution.*[6]

7.19 *Boric Acid*—Solid.

7.20 *Isopropyl Ether.*

8. Sampling

8.1 Collect the sample in accordance with Practice C998.

8.2 Prepare the sample for analysis in accordance with Practice C999.

9. Calibration and Standardization

9.1 If a standard uranium-232 solution is not available for use as a tracer, standardize a freshly prepared sample of uranium-232; for guidance refer to Practices D3648. This standard may also be used to establish the counting efficiency of the alpha spectrometer which then can be used to calculate the chemical yield for each sample.

10. Precautions

10.1 Adequate laboratory facilities, such as fume hoods and controlled ventilation, along with safe techniques, must be used

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[4] *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. Pharmacopeial Convention, Inc. (USPC), Rockville, MD.

[5] Ag1-X8 from BioRad Laboratories (Hercules, CA) and A8-B500-M-Cl from Eichrom Technoloigies, Inc.(Lisle, IL) have been found to be satisfactory.

[6] Uranium-232 is available from the National Institute of Standards and Technology, Gaithersburg, MD 20899, as a Standard Reference Material.

in this procedure. Extreme care should be exercised in using hydrofluoric and other hot, concentrated acids. Use of proper gloves is recommended. Refer to the laboratory's chemical hygiene plan and other applicable guidance for handling chemical and radioactive materials and for the management of radioactive, mixed, and hazardous waste.

10.2 Hydrofluoric acid is a highly corrosive acid that can severely burn skin, eyes, and mucous membranes. Hydrofluoric acid is similar to other acids in that the initial extent of a burn depends on the concentration, the temperature, and the duration of contact with the acid. Hydrofluoric acid differs from other acids because the fluoride ion readily penetrates the skin, causing destruction of deep tissue layers. Unlike other acids that are rapidly neutralized, hydrofluoric acid reactions with tissue may continue for days if left untreated. Due to the serious consequences of hydrofluoric acid burns, prevention of exposure or injury of personnel is the primary goal. Utilization of appropriate laboratory controls (hoods) and wearing adequate personal protective equipment to protect from skin and eye contact is essential.

11. Procedure

11.1 *Acid Dissolution:*

11.1.1 Weigh a 10.0 ± 0.1 g soil sample into a porcelain crucible.

11.1.2 Add an appropriate amount of uranium-232 tracer to the sample. (If the activity of the sample is expected to be less than 0.01 Bq/g or is unknown, add 0.1 Bq of tracer. For higher levels add uranium-232 tracer which is equivalent to the estimated activity of uranium in the sample.)

11.1.3 Heat the porcelain crucible containing the soil sample in a muffle furnace at 600°C for 4 h, remove, and cool.

11.1.4 Transfer the sample to a 250-mL TFE-fluorocarbon beaker and rinse the porcelain crucible several times with concentrated HNO_3 until the final volume is 60 mL.

11.1.5 Add 30 mL of 48 % HF to the beaker and cover with a TFE-fluorocarbon watchglass. Heat the sample for one hour and stir frequently with a TFE-fluorocarbon stirring rod during the heating. Cool the solution.

11.1.6 Add 30 mL each of concentrated HNO_3 and 48 % HF and digest with some stirring for an additional hour.

11.1.7 Add 20 mL of concentrated HCl and heat. Occasionally stir the solution.

11.1.8 Remove the cover and evaporate the solution to approximately 20 mL. Cool the solution.

11.1.9 Add 50 mL of water and 5 g boric acid to the solution and heat while stirring for 10 min to dissolve the soluble salts.

11.1.10 Cool and transfer the sample solution to a 250-mL centrifuge bottle and wash the beaker with a minimum amount of water and combine.

11.1.11 Centrifuge and transfer the supernate to a 250-mL centrifuge bottle.

11.1.12 Wash the residue with 10 mL of 1 M HCl, centrifuge, and add the washing to the supernate.

11.2 *Co-Precipitation:*

11.2.1 Add 2 mL of 0.18 M ferric chloride solution to the supernate in the centrifuge bottle and stir. It may not be necessary to add the iron carrier if a sufficient amount of iron is present in the soil.

11.2.2 Add concentrated NH_4OH to the sample solution while mixing to precipitate the iron and until the pH reaches 9 to 10.

11.2.3 Add 5 mL of concentrated NH_4OH in excess.

11.2.4 Centrifuge the sample for 5 min, discard the supernate. Dissolve the precipitate with a minimum addition of concentrated HCl and bring the volume to 50 mL with 8 M HCl.

11.2.5 Transfer the sample to a 250-mL separatory funnel and rinse the centrifuge bottle with two 5 mL rinses of 8 M HCl.

11.3 *Ether Extraction:*

11.3.1 Add 60 mL of isopropyl ether to the funnel, stopper, and shake for 2 min. Allow the phases to separate and drain the aqueous phase into a second separatory funnel.

11.3.2 Add 5 mL of concentrated HCl to the second funnel and mix. Add 60 mL of isopropyl ether and repeat the extraction twice more to remove iron. (Additional extractions may be necessary if the third extract is yellow, indicating incomplete removal of the iron).

11.3.3 Transfer the aqueous phase to a 150-mL beaker and boil the solution for 15 min.

11.4 *Anion Exchange Separation:*

11.4.1 Condition the anion exchange column (7.7) by rinsing with four column volumes of 8 M HCl.

11.4.2 Transfer the sample to the anion exchange column and pass the sample through the column at a rate of 2 mL per min.

11.4.3 Wash the column with six column volumes of 6 M HCl containing 1 mL of concentrated HI per 50 mL of 6 M HCl, prepared immediately before use, to remove iron and plutonium.

11.4.4 Wash the column with two column volumes of 6 M HCl.

11.4.5 Elute the uranium with six column volumes of 1 M HCl and collect in a 150-mL beaker.

11.4.6 Evaporate the sample to 20 mL and add 5 mL of concentrated HNO_3. Evaporate the sample to near dryness.

11.5 *Electrodeposition:*

11.5.1 Uranium can also be prepared for alpha spectrometry by coprecipitation with neodymium fluoride, see Practice C1163 or by electrodeposition using Practice C1284.

11.5.2 Count the sample on a calibrated alpha pulse height analyses system within 1 week or sooner to prevent interferences from uranium-232 daughters.

11.6 *Alpha Pulse Height Analysis:*

11.6.1 Count the sample on an alpha spectrometer for 1000 min or longer to resolve the uranium isotopes.[2]

11.6.2 Determine the background and reagent blank activities and correct the count for each peak.

TABLE 1 Summary of Uranium Results

Isotope	Reference Value (Bq/g)	Number of Laboratories	$\bar{X}$	Percent Relative Standard Deviation	
				Within Laboratory	Between Laboratories
^{238}U	0.0035 ± 0.0005	4	0.0032	26.3	33.0
	0.032 ± 0.001	7	0.031	8.9	...
	0.378 ± 0.027	5	0.36	8.8	4.6
	0.342 ± 0.0038	3	0.35	4.8	1.0
^{234}U	0.004 ± 0.0007	4	0.0042	44.0	64.0
	0.046 ± 0.0013	7	0.0447	10.0	2.6
	0.342 ± 0.0038	3	0.344	4.5	...
	0.730 ± 0.0057	5	0.632	16.0	9.8
^{235}U	0.0016± 0.0001	5	0.0015	11.0	16.0
	0.0148 ± 0.0003	2	0.017	6.0	37.0
	0.0317 ± 0.0012	4	0.028	16.0	21.0
^{236}U	0.022 ± 0.0005	5	0.022	12.0	...

12. Calculation

12.1 The activity of each uranium isotope is calculated as follows:

$$X_i = (C_iA_t)/(C_tW_s) \tag{1}$$

where:

X_i = concentration of a uranium isotope in Bq/g,
A_t = activity of the yield tracer added in Bq,
C_i = net sample counts in the energy region of the uranium isotope being measured,
C_t = net sample counts in the energy region of the uranium-232 tracer, and
W_s = the sample weight in g.

12.2 The fractional counting efficiency of the alpha spectrometer, *E*, must be determined if it is desired to calculate the chemical yield for uranium. Calculate the fractional efficiency as follows:

$$E = R_s/A_{std} \tag{2}$$

where:

R_s = net counting rate of the standard source in the region of the alpha emitter(s) of interest in cps, and
A_{std} = activity of the standard source in Bq at the time of the count.

12.3 Calculate the fractional chemical yield for uranium as follows:

$$Y = (C_t)/(TA_tE) \tag{3}$$

where:

T = counting time in s,
C_t = the net sample counts in the energy region of the uranium-232 tracer,
A_t = activity of the yield tracer added in Bq, and
E = fractional counting efficiency of the alpha spectrometer.

12.4 Refer to Practice D3084 for the Minimum Detectable Concentration (MDC), critical level, and uncertainty calculations.

13. Precision and Bias

13.1 *Precision*—Reference samples were sent to fourteen laboratories. Analytical data on some or all of the samples was returned by eight of the laboratories.[2] It is not known if these eight laboratories are typical of the laboratories that will use this test method. Table 1 summarizes the results from these laboratories.

13.2 *Bias*—There is no indication of bias over the samples. In all cases except one (^{235}U in Samples 2 and 3), the computed $\bar{X}$ was within the two standard deviation limits on the reference value. For the Sample 3 cases, the uncertainty on the computed $\bar{X}$ was so large, the reference value was covered.

14. Keywords

14.1 alpha spectrometry; radiochemistry; soil; uranium

APPENDIX

(Nonmandatory Information)

X1. ALPHA ENERGIES FOR ISOTOPES OF URANIUM

TABLE X1.1 Properties of Uranium Isotopes of Interest in Environmental Samples[A]

Isotope	Half-Life (years)	Alpha Energies, MeV (abundance)
^{232}U	68.9	5.320 (68.6 %), 5.263 (31.2 %)
^{233}U	1.592×10^5	4.825 (84.4 %), 4.783 (13.2 %)
^{234}U	2.454×10^5	4.776 (72.5 %), 4.723 (27.5 %)
^{235}U	7.037×10^8	4.395 (85 %), 4.370 (6 %), 4.597, (5 %)
^{236}U	2.342×10^7	4.494 (74 %), 4.445 (26 %)
^{238}U	4.468×10^9	4.196 (77 %), 4.147 (23 %)

[A] Available from Browne, E. and Firestone, R. B., *Table of Radioactive Isotopes*, (Shirley, V. S., Ed.), John Wiley and Sons, Inc., 1986.

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