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# **Improved Rapid Radiochemical Method for Radium-226 in Building Materials for Environmental Remediation Following Radiological Incidents**

**U.S. Environmental Protection Agency**

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**Improved Rapid Radiochemical Method for Radium-226 in Building Materials for  
Environmental Remediation Following Radiological Incidents using DGA Resin**

**Revision History**

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**IMPROVED RAPID RADIOCHEMICAL METHOD FOR RADIUM-226 IN BUILDING MATERIALS FOR ENVIRONMENTAL REMEDIATION FOLLOWING RADIOLOGICAL INCIDENTS**

1. Scope and Application

- 1.1. The method will be applicable to samples where contamination is either from known or unknown origins.
- 1.2. This method uses improved rapid radiochemical separations techniques for the isotopic determination of  $^{226}\text{Ra}$  in building materials such as asphalt, shingles, limestone and granite following a nuclear or radiological incident. The method has been validated for asphalt and asphalt shingles. The matrix specific digestion procedures digest the samples, removing organics if necessary, rapidly pre-concentrating  $^{226}\text{Ra}$  from the digested samples so that this method can purify this alpha-emitting isotope for measurement.
- 1.3. This method is a shorter, newer alternative to *Rapid Radiochemical Method for Radium-226 in Brick and Concrete for Environmental Remediation Following Radiological Incidents* (Reference 16.5). It combines the final purification of Ra-226 into a single column (Sr Resin + DGA Resin) instead of two separate steps involving Sr Resin and then Ln Resin. Other solid samples such as soil, brick and concrete can be digested using the rapid sodium hydroxide fusion procedure as an alternative to other digestion techniques, but application of this  $^{226}\text{Ra}$  procedure to these samples will have to be validated by the laboratory.
- 1.4. This method is specific for  $^{226}\text{Ra}$ . It uses 50WX8 cation resin to separate radium from solid matrix constituents, followed by additional separation steps using Sr Resin and DGA Resin to remove interferences.
- 1.5. The method is capable of satisfying the following required method uncertainty for  $^{226}\text{Ra}$  in each respective matrix. To attain the stated measurement quality objectives (MQOs), a sample aliquant of approximately 1 g and count time of 8 hours (or longer) are recommended. Application of the method must be validated by the laboratory using the protocols provided in *Method Validation Guide for Qualifying Methods Used by Radiological Laboratories Participating in Incident Response Activities* (Reference 16.1). The sample turnaround time and throughput may vary based on additional project MQOs, the time for analysis of the sample test source, and initial sample weight/volume.
  - 1.5.1. 0.34 pCi/g at an analytical action level of 2.61 pCi/g in asphalt shingles
  - 1.5.2. 0.67 pCi/g at an analytical action level of 5.16 pCi/g in asphalt
  - 1.5.3. 0.62 pCi/g at an analytical action level of 4.75 pCi/g in limestone
- 1.6. The rapid  $^{226}\text{Ra}$  method was evaluated following the guidance presented for “Level E Method Validation: Adapted or Newly Developed Methods, Including Rapid Methods” in *Method Validation Guide for Qualifying Methods Used by Radiological Laboratories Participating in Incident Response Activities* (Reference 16.1) and Chapter 6 of *Multi-Agency Radiological Laboratory Analytical Protocols Manual* (Reference 16.2).

## 2. Summary of Method

- 2.1. A known quantity of  $^{225}\text{Ra}$  is used as the yield tracer in this analysis. The sample is fused using a digestion procedure specific for the solid sample analyzed.
- 2.2. The method has been initially validated using pulverized asphalt shingles which are digested using *Rapid Method for Sodium Hydroxide Fusion of Asphalt Roofing Materials Matrices Prior to Americium, Plutonium, Strontium, Radium, and Uranium Analyses* (Reference 16.6) and pulverized asphalt which are digested using *Rapid Method for Sodium Hydroxide Fusion of Asphalt Matrices Prior to Americium, Plutonium, Strontium, Radium, and Uranium Analyses* (Reference 16.7). This method has been applied to the  $^{226}\text{Ra}$  method validation process for the limestone matrix. Alternate matrices must be digested per the appropriate digestion procedure and validated by the laboratory.
- 2.3. Radium isotopes are removed from the fusion matrix using a carbonate precipitation step. The sample is acidified and loaded onto 50WX8 cation resin to remove sample interferences such as calcium. The radium is eluted from the cation resin with 8M nitric acid. After evaporation of the eluate, the sample is dissolved and passed through a stacked Sr Resin +DGA Resin column. Ba is removed using Sr Resin while DGA Resin is used to remove interferences such as residual calcium and to remove the initial  $^{225}\text{Ac}$  present. The purified eluent is evaporated to dryness and redissolved in 1.5M HCl. The radium (including  $^{226}\text{Ra}$ ) is prepared for counting by microprecipitation with  $\text{BaSO}_4$ .
- 2.4. Low-level measurements are performed by alpha spectrometry. The activity measured in the  $^{226}\text{Ra}$  region of interest is corrected for chemical yield based on the observed activity of the alpha peak at 7.07 MeV ( $^{217}\text{At}$ , the third progeny of  $^{225}\text{Ra}$ ). See Table 17.1 for a list of alpha particle energies of the radionuclides that potentially may be seen in the alpha spectra.

## 3. Definitions, Abbreviations and Acronyms

- 3.1. Analytical Protocol Specifications (APS). The output of a *directed planning process* that contains the project's analytical data needs and requirements in an organized, concise form.
- 3.2. Analytical Action Level (AAL). The term "analytical action level" is used to denote the value of a quantity that will cause the decisionmaker to choose one of the alternative actions.
- 3.3. Discrete Radioactive Particles (DRPs or "hot particles"). Particulate matter in a sample of any matrix where a high concentration of radioactive material is present as a very small particle (<millimeter [mm]).
- 3.4. *Multi-Agency Radiological Analytical Laboratory Protocols Manual* (MARLAP) provides guidance for the planning, implementation, and assessment phases of those projects that require the laboratory analysis of radionuclides (Reference 16.1).
- 3.5. Measurement Quality Objective (MQO). The analytical data requirements of the data quality objectives that are project- or program-specific and can be quantitative or qualitative. These analytical data requirements serve as measurement performance criteria or objectives of the analytical process.

- 3.6. Radiological Dispersal Device (RDD), i.e., a “dirty bomb.” This device is an unconventional weapon constructed to distribute radioactive material(s) into the environment either by incorporating them into a conventional bomb or by using sprays, canisters, or manual dispersal.
  - 3.7. Required Method Uncertainty ( $u_{MR}$ ). The required method uncertainty is a target value for the individual measurement uncertainties and is an estimate of uncertainty (of measurement) before the sample is actually measured. The required method uncertainty as an absolute value is applicable at or below an AAL.
  - 3.8. Relative Required Method Uncertainty ( $\phi_{MR}$ ). The relative required method uncertainty is the  $u_{MR}$  divided by the AAL and is typically expressed as a percentage. It is applicable above the AAL.
  - 3.9. Sample Test Source (STS). This is the final form of the sample that is used for nuclear counting. This form is usually specific for the nuclear counting technique in the method, such as a solid deposited on a filter for alpha spectrometry analysis.
4. Interferences
    - 4.1. Radiological
      - 4.1.1. Unless other radium isotopes are present in concentrations greater than approximately three times the  $^{226}\text{Ra}$  activity concentration, interference from other radium alphas will be resolved when using alpha spectrometry. Method performance may be compromised if samples contain high levels of radium isotopes due to ingrowth of interfering decay progeny, but this will depend on the actual spectral resolution.
      - 4.1.2. Radionuclides with overlapping alpha energies such as  $^{229}\text{Th}$ ,  $^{234}\text{U}$ , and neptunium-237 ( $^{237}\text{Np}$ ) will interfere if they are not removed effectively. The method removes these radionuclides.
      - 4.1.3. Decay progeny from the  $^{225}\text{Ra}$  tracer will continue to ingrow as more time elapses between the separation of radium and the count of the sample. Delaying the count significantly longer than a day may introduce a possible positive bias in results near the detection threshold. When MQOs require measurements close to detection levels, and coordinating sample processing and counting schedules is not conducive to counting the sample within approximately 36 hours of the separation of radium, the impact of tracer progeny tailing into the  $^{226}\text{Ra}$  may be minimized by reducing the activity of the  $^{225}\text{Ra}$  tracer that is added to the sample. This will aid in improving the signal-to-noise ratio for the  $^{226}\text{Ra}$  peak by minimizing the amount of tailing from higher energy alphas of the  $^{225}\text{Ra}$  progeny.
      - 4.1.4. There is also a possibility that the higher energy peaks associated with the  $^{225}\text{Ra}$  progeny may result in energy-attenuated counts that show up in the lower energy  $^{226}\text{Ra}$  alpha spectra region so reducing the  $^{225}\text{Ra}$  tracer while still achieving enough  $^{217}\text{At}$  counts to minimize tracer uncertainty may be optimal.
        - 4.1.4.1. The amount of  $^{225}\text{Ra}$  added to the samples may be decreased, and the time for ingrowth between separation and counting increased,

to ensure that sufficient  $^{225}\text{Ac}$ , francium-221 ( $^{221}\text{Fr}$ ), and  $^{217}\text{At}$  are present for yield corrections at the point of the count. Although this detracts from the rapidity of the method, it does not detract significantly from the potential for high throughput.

- 4.1.5. A purified  $^{225}\text{Ra}$  tracer solution may be used when performing this method (see Appendix that follows this method).
  - 4.1.5.1. When using a purified source of  $^{225}\text{Ra}$ , the beginning of decay for  $^{225}\text{Ra}$  is the activity reference date established during standardization of the  $^{225}\text{Ra}$  solution.
- 4.1.6. It is also possible to use  $^{225}\text{Ra}$  in equilibrium with thorium-229 ( $^{229}\text{Th}$ ) for convenience, which may be added to each sample as a tracer.<sup>1</sup> This allows use of  $^{229}\text{Th}$  without purification and therefore is a simpler approach. This approach requires complete decontamination of a relatively high activity of  $^{229}\text{Th}$  in the later steps in the method, since the spectral region of interest (ROI) for  $^{229}\text{Th}$  slightly overlaps that of  $^{226}\text{Ra}$ .
- 4.1.7.  $^{229}\text{Th}$  is removed during the cation exchange step (retained) and the  $^{225}\text{Ra}$  is unsupported from this point on in the method (retained on the cation resin). If the time delay between the cation exchange step and the DGA Resin separation of  $^{229}\text{Th}$  is 6 hours or less the error associated with the  $^{225}\text{Ra}$  reference value is  $\leq 1.2\%$  due to  $^{225}\text{Ra}$  decay. A correction for this decay can also be made by recording the cation exchange elution time, and decaying  $^{225}\text{Ra}$  from this point until the DGA Resin separation time to eliminate this relatively small bias.
- 4.1.8. The method provides effective removal of  $^{229}\text{Th}$ . Inadequate decontamination of  $^{229}\text{Th}$  may lead to high bias in the  $^{226}\text{Ra}$  result especially when the levels of  $^{226}\text{Ra}$  in the sample are below 1 pCi/g. The spectral region above  $^{226}\text{Ra}$  corresponding to  $^{229}\text{Th}$  should be monitored routinely to identify samples where  $^{229}\text{Th}$  interference may impact compliance with project MQOs. If problematic levels of  $^{229}\text{Th}$  are identified in spectra, measures must be taken to address the interference. These might include:
  - 4.1.8.1. Separating  $^{225}\text{Ra}$  from  $^{229}\text{Th}$  prior to its use as a tracer.
  - 4.1.8.2. Increasing the sample aliquant size without changing the amount of tracer added will increase the analyte signal and reduce the relative impact of the interference to levels that may be amenable with project MQOs.
  - 4.1.8.3. The absolute amount of  $^{229}\text{Th}$  added to the samples may be decreased, as long as the time for ingrowth between separation and counting is increased to ensure that sufficient  $^{217}\text{At}$  is present for yield corrections at the point of the count. Although this detracts from the rapidity of the method, it allows more

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<sup>1</sup> The single-laboratory validation for this method was performed successfully by adding  $^{225}\text{Ra}$  in secular equilibrium with  $^{229}\text{Th}$  tracer. See the Appendix of this method for a method for separating (and standardizing)  $^{225}\text{Ra}$  tracer from  $^{229}\text{Th}$  solution.

flexibility in the timing of the count and does not detract from the potential for high throughput.

- 4.1.8.4. The samples may be counted as early as about 8 hours after separation time with an 8-hour count time if ~100 pCi  $^{229}\text{Th}$  is added, but separation times and counting time midpoints must be recorded carefully and precisely.
- 4.1.9. When a solution containing  $^{225}\text{Ra}$  in equilibrium with  $^{229}\text{Th}$  is used as a tracer, thorium is removed during the processing of the sample. The equilibrium between the  $^{225}\text{Ra}$  and  $^{229}\text{Th}$  is essentially maintained until the cation exchange elution step is performed. At this point, the  $^{225}\text{Ra}$  activity in the eluate is unsupported and begins to decay.  $^{225}\text{Ac}$  is removed during the DGA Resin separation.
- 4.1.10. Ascorbic acid is added to the sample load solution to reduce  $\text{Fe}^{3+}$  present to  $\text{Fe}^{2+}$ , which has less retention on cation resin than  $\text{Fe}^{3+}$ .
- 4.1.11. Trace levels of  $^{226}\text{Ra}$  may be present in  $\text{Na}_2\text{CO}_3$  used in the  $^{226}\text{Ra}$  pre-concentration step of the fusion method. Adding less 2M  $\text{Na}_2\text{CO}_3$  (<10 mL used in this method) may reduce  $^{226}\text{Ra}$  reagent blank levels, while still effectively pre-concentrating  $^{226}\text{Ra}$  from the fusion matrix. This will need to be validated by the laboratory.
- 4.2. Non-radiological
  - 4.2.1. The amount of inherent stable (non-radioactive) barium in the sample that may be carried through the processes prior to microprecipitation should not significantly exceed the amount of the barium carrier (50  $\mu\text{g}$ ), which is added for microprecipitation. Microprecipitates on the STS greater than 50  $\mu\text{g}$  Ba may degrade the resolution of alpha spectra.
    - 4.2.1.1. In this procedure, Ba is removed using Sr Resin and alpha peak resolution is typically very good. It is important for the total volume of 4M  $\text{HNO}_3$  passed through Sr Resin to be kept relatively small per procedure to remove Ba effectively. It is likely that Sr Resin can be washed and reused to reduce resin costs, but this will have to be validated by the laboratory.
    - 4.2.1.2. The removal of Ba allows a larger aliquant size of samples to be analyzed that could not typically be tolerated in methods that do not remove Ba, allowing shorter count times and lower minimum detectable activity levels.
  - 4.2.2. Ca can also cause alpha peak resolution problems and needs to be effectively removed. Most of the Ca ions are removed using the initial cation exchange separation. A small amount is removed during the final DGA Resin purification step.
  - 4.2.3. A smaller sample size may need to be selected when these interferences cannot be removed adequately.
  - 4.2.4. The DGA Resin step provides a final purification for the Ra-225 tracer. If the flow rate is too fast (>1.5 drops/second) and Ac-225 is present prior to the

final separation time breaks through the resin, a high bias in the tracer yield will occur.

## 5. Safety

### 5.1. General

- 5.1.1. Refer to your safety manual for concerns of contamination control, personal exposure monitoring and radiation dose monitoring.
- 5.1.2. Refer to your laboratory's chemical hygiene plan for general chemical safety rules.

### 5.2. Radiological

#### 5.2.1. Hot Particles (DRPs)

- 5.2.1.1. Hot particles, also termed "discrete radioactive particles" (DRPs), will likely be small, on the order of 1 mm or less. DRPs typically are not evenly distributed in the media and their radiation emissions are not uniform in all directions (anisotropic).

- 5.2.2. For samples with detectable activity concentrations of these radionuclides, labware should be used only once due to the potential for cross contamination.

### 5.3. Procedure-Specific Non-Radiological Hazards:

- 5.3.1. Solutions of 30% H<sub>2</sub>O<sub>2</sub> can rapidly oxidize organic materials and generate significant heat. Do not mix large quantities of peroxide solution with solutions of organic solvents as the potential for conflagration exists.

## 6. Equipment and supplies

- 6.1. Alpha spectrometer calibrated for use over the range of ~3.5-10 MeV.
- 6.2. Cartridge reservoirs, 10 or 20 mL syringe style with locking device, or reservoir columns (empty luer tip, CC-10-M) plus 12 mL reservoirs (CC-06-M), Image Molding, Denver, Colorado, or equivalent.
- 6.3. Centrifuge tubes, polypropylene, 50 mL, disposable; or equivalent.
- 6.4. Chromatography columns, polypropylene, disposable:
  - 6.4.1. 1.5 centimeter (cm) inner diameter × 15 cm; or equivalent (Environmental Express, Mount Pleasant, South Carolina).
  - 6.4.2. Additional frits for 1.5 cm inner diameter × 15 cm columns (Environmental Express, Mount Pleasant, South Carolina).
- 6.5. Filter funnels.
- 6.6. Filter manifold apparatus with 25 mm diameter polysulfone. A single-use (disposable) filter funnel/filter combination may be used, to avoid cross-contamination.
- 6.7. 100 µL, 200 µL, 500 µL and 1 mL pipets or equivalent and appropriate plastic tips.
- 6.8. 1-10 mL electronic pipet or manual equivalent.



- 6.9. Glass beaker, 50 mL, 150 mL and 250 mL capacity.
- 6.10. Heat lamp.
- 6.11. Hot plate.
- 6.12. Graduated cylinders, 500 mL and 1000 mL.
- 6.13. 25 mm polypropylene filter, 0.1  $\mu\text{m}$  pore size, or equivalent.
- 6.14. pH paper.
- 6.15. Stainless steel planchets or other adhesive sample mounts (Ex. Environmental Express, Inc. P/N R2200) able to hold the 25 mm filter.
- 6.16. Tips, white inner, Eichrom part number AC-1000-IT, or PFA 5/32"  $\times$  1/4" heavywall tubing connectors, natural, Ref P/N 00070EE, cut to 1 inch, Cole Parmer, or equivalent
- 6.17. Tips, yellow outer, Eichrom part number AC-1000-OT, or equivalent.
- 6.18. Tweezers.
- 6.19. Vacuum box, such as Eichrom part number AC-24-BOX, or equivalent.
- 6.20. Vacuum pump or laboratory vacuum system.
- 6.21. Vortex mixer.
- 6.22. Heat lamp.

## 7. Reagents and Standards

### NOTES:

**All reagents are American Chemical Society reagent grade or equivalent unless otherwise specified.**

**Unless otherwise indicated, all references to water should be understood to mean Type I reagent water (ASTM D1193, Reference 16.3). For microprecipitation, all solutions used in microprecipitation should be prepared with water filtered through a 0.45  $\mu\text{m}$  (or smaller) filter.**

- 7.1. Type I reagent water as defined in ASTM Standard D1193 (Reference 16.3).
- 7.2. Ammonium sulfate, solid  $(\text{NH}_4)_2\text{SO}_4$ .
- 7.3. Barium carrier (1000  $\mu\text{g}/\text{mL}$  as  $\text{Ba}^{2+}$ ). May be purchased as an inductively coupled plasma – atomic emission spectrometry (ICP-AES) standard and diluted, or prepared by dissolving 0.90 g reagent grade barium chloride, dihydrate ( $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$ ) in water and diluting to 500 mL with water.
- 7.4. Calcium nitrate (1.25M): Dissolve 147 g of calcium nitrate tetrahydrate ( $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ ) in 300 mL of water and dilute to 500 mL with water.
- 7.5. Cation Resin, 50WX8, 200–400  $\mu\text{m}$  mesh size [available from Eichrom Technologies, Lisle, IL].
- 7.6. DGA Resin cartridges, 2 mL, small particle size (50–100  $\mu\text{m}$ ), in appropriately sized column pre-packed cartridges.
- 7.7. Ethanol, reagent 95% ( $\text{C}_2\text{H}_5\text{OH}$ ), available commercially.
- 7.8. Hydrochloric acid (12M): Concentrated HCl, available commercially.
  - 7.8.1. Hydrochloric acid (6.0M): Add 500 mL of concentrated HCl to 300 mL of water and dilute to 1.0 L with water.

- 7.8.2. Hydrochloric acid (3.0M): Add 250 mL of concentrated HCl to 600 mL of water and dilute to 1.0 L with water.
  - 7.8.3. Hydrochloric acid (1.5M): Add 125 mL of concentrated HCl to 800 mL of water and dilute to 1.0 L with water.
  - 7.8.4. Hydrochloric acid (0.5M): Add 41.5 mL of concentrated HCl to 800 mL of water and dilute to 1.0 L with water.
  - 7.9. Hydrogen peroxide, H<sub>2</sub>O<sub>2</sub> (30 % weight/weight), available commercially.
  - 7.10. Isopropanol, 2-propanol, (C<sub>3</sub>H<sub>7</sub>OH), available commercially.
    - 7.10.1. Isopropanol (2-propanol), 20% (volume/) in water: Mix 20 mL of isopropanol with 80 mL of water.
  - 7.11. Methanol (CH<sub>3</sub>OH), available commercially
  - 7.12. Nitric acid (16M): Concentrated HNO<sub>3</sub>, available commercially.
    - 7.12.1. Nitric acid (8M): Add 506 mL of concentrated HNO<sub>3</sub> to 300 mL of water and dilute to 1.0 L with water.
    - 7.12.2. Nitric acid (4M): Add 253 mL of concentrated HNO<sub>3</sub> to 200 mL of water and dilute to 1.0 L with water.
  - 7.13. Ra-225 tracer in 1M HCl solution in a concentration amenable to accurate addition of about 180 dpm per sample (generally about 150–600 dpm/mL).
    - 7.13.1. Ra-225 may be purified and standardized using a <sup>229</sup>Th/<sup>225</sup>Ra generator as described in the appendix of this method.
    - 7.13.2. Th-229 (~70-100 pCi) containing an equilibrium concentration of <sup>225</sup>Ra has been successfully used without prior separation of the <sup>225</sup>Ra.
  - 7.14. Sr Resin cartridges, 2 mL, small particle size (50–100 μm), in appropriately sized column pre-packed cartridges.
8. Sample Collection, Preservation, and Storage  
Not Applicable.
9. Quality Control
- 9.1. Batch quality control results shall be evaluated and meet applicable Analytical Protocol Specifications (APS) prior to release of unqualified data. In the absence of project-defined APS or a project-specific quality assurance project plan (QAPP), the quality control sample acceptance criteria defined in the laboratory quality manual and procedures shall be used to determine acceptable performance for this method.
    - 9.1.1. A laboratory control sample (LCS) shall be run with each batch of samples. The concentration of the LCS should be at or near the action level or a level of interest for the project.
    - 9.1.2. One method blank shall be run with each batch of samples. The laboratory blank should consist of an acceptable simulant or empty crucible blank processed through the fusion procedure. If an empty crucible is used to generate a reagent blank sample, it is recommended that 150 mg Ca be

added as calcium nitrate to the empty crucible as blank simulant. This facilitates Ra carbonate precipitations from the alkaline fusion matrix.

- 9.1.3. One laboratory duplicate shall be run with each batch of samples. The laboratory duplicate is prepared by removing an aliquant from the original sample container.
- 9.1.4. A matrix spike sample may be included as a batch quality control sample if there is concern that matrix interferences, such as the presence of elemental barium in the sample, may compromise chemical yield measurements, or overall data quality.
- 9.2. Sample-specific quality control measures
  - 9.2.1. Limits and evaluation criteria shall be established to monitor each alpha spectrum to ensure that spectral resolution and peak separation is adequate to provide quantitative results. When  $^{229}\text{Th}/^{225}\text{Ra}$  solution is added directly to the sample, the presence of detectable counts between ~5.0 MeV and the upper boundary established for the  $^{226}\text{Ra}$  ROI generally indicates the presence of  $^{229}\text{Th}$  in the sample, and in the  $^{226}\text{Ra}$  ROI. If the presence of  $^{229}\text{Th}$  is noted and the concentration of  $^{226}\text{Ra}$  is determined to be an order of magnitude below the action limit or the detection threshold of the method, take corrective actions to ensure that MQOs have not been compromised (e.g., clean up  $^{225}\text{Ra}$  tracer before adding, or re-process affected samples and associated QC samples. See interferences sections Steps 4.1.4 – 4.1.5. for discussion).
- 9.3. This method is capable of achieving a  $u_{\text{MR}}$  of 0.34 and 0.83 pCi/g at or below an action level of 6.41 pCi/g for shingles and a  $u_{\text{MR}}$  of 0.34 pCi/g at or below an action level of 2.61 pCi/g for asphalt. This may be adjusted in the event specific MQOs are different. .
- 9.4. This method is capable of achieving a  $\phi_{\text{MR}}$  13% above 6.41 pCi/g for shingles and 2.61 pCi/g for asphalt. This may be adjusted if the event specific MQOs are different.
- 9.5. This method is capable of achieving a required minimum detectable concentration (MDC) of ~10.5 pCi/g in a counting time of six hours for shingles and ~0.21 pCi/g for asphalt.

## 10. Calibration and Standardization

- 10.1. Set up, operate, calibrate and perform quality control for alpha spectrometry units in accordance with the laboratory's quality manual and standard operating procedures and consistent with ASTM Standard Practice D7282, Sections 7-13, 18, and 24 (Reference 16.4).

**NOTE: The calibrated energy range for the alpha spectrometer for this method should be from ~3.5 to 10 MeV.**
- 10.2. If  $^{225}\text{Ra}$  is separated and purified from  $^{229}\text{Th}$  for use as a tracer, the activity reference date established during standardization of the tracer is used as the  $^{225}\text{Ra}$  activity reference date. (See the appendix of this method.)
- 10.3. When using  $^{229}\text{Th}$  containing an equilibrium concentration of  $^{225}\text{Ra}$ , the time of most recent separation/purification of the  $^{229}\text{Th}$  standard solution must be known in order

to determine the extent of secular equilibrium between  $^{229}\text{Th}$  and its  $^{225}\text{Ra}$  progeny. Verify the date of purification by examining the Certificate of Analysis, or other applicable documentation, for the standard.

- 10.4. When using  $^{229}\text{Th}$  containing an equilibrium concentration of  $^{225}\text{Ra}$ ,  $^{225}\text{Ra}$  is separated from its  $^{229}\text{Th}$  parent in the solution during the cation exchange elution step. This is the beginning of  $^{225}\text{Ra}$  decay and the date and time used for decay correction of the tracer. This time must be known and recorded precisely.
  - 10.4.1. If the purification date of the  $^{229}\text{Th}$  is not documented, at least 100 days must have elapsed between separation and use to ensure that  $^{229}\text{Th}$ , and its progeny  $^{225}\text{Ra}$  are in full secular equilibrium (i.e., >99%. Table 17.3).

## 11. Procedure

### 11.1. Initial Sample Preparation for Radium

- 11.1.1. Ra isotopes are preconcentrated from solid samples using the appropriate sodium hydroxide (NaOH) fusion procedure, such as *Rapid Method for Sodium Hydroxide Fusion of Asphalt Roofing Material Matrices Prior to Americium, Plutonium, Strontium, Radium, and Uranium Analyses* (Reference 16.6), which fuses the samples using rapid NaOH fusion followed by carbonate precipitation to preconcentrate Ra isotopes from the hydroxide matrix. Alternate digestion methods specific to the solid matrix may also be used, but this will have to be validated by the laboratory.
- 11.1.2. The carbonate precipitate is dissolved in an HCl solution and additional separation steps to purify the radium isotopes are performed using this procedure.
- 11.1.3. A smaller volume of the total load solution may be taken and analyzed as needed for very high activity samples, with appropriate dilution factor calculations applied.
- 11.1.4. This separation can be used with other solid sample matrices dissolved in 0.1M to 1.5M HCl.

### 11.2. Initial Matrix Removal Using 50WX8 Cation Resin

#### 11.2.1. Prepare sample solution

- 11.2.1.1. Add 3 mL of 1.5M ascorbic acid to each sample solution to reduce any Fe present to  $\text{Fe}^{2+}$ . Mix and wait approximately 3 minutes.

#### 11.2.2. Set up vacuum box

**NOTE: More than one vacuum box may be used to increase throughput as needed.**

- 11.2.2.1. For each sample solution, place the empty large columns (15 cm columns or equivalent) on the vacuum box.
- 11.2.2.2. Add a water slurry (or weigh out the solid resin) of cation resin 50WX8 (200-400 mesh) into each column equivalent to 5 g of resin.
- 11.2.2.3. Turn the vacuum on and ensure proper fitting of the lid.

**IMPORTANT: The unused openings on the vacuum box should be sealed. Yellow caps (included with the vacuum box) can be used to plug unused white tips to achieve good seal during the separation. Alternately, plastic tape can be used to seal the unused lid holes as well.**

- 11.2.2.4. After the water has passed through, place a frit down on top of the resin bed.
- 11.2.2.5. Add additional water (~10–15 mL) to rinse the resin and remove fine resin particles. Clean resin by adding 15 mL 6M HCl and allow to drain at ~2 mL/min, then 15 ml water ~2 mL/min to rinse each column.
- 11.2.2.6. Add 10 mL of 0.5M HCl to the column to precondition the resin.
- 11.2.2.7. Press frit down tightly on resin bed.  
**NOTE: It is important to control flow rates such that they are not too fast. Gravity flow (no vacuum) may be adequate, although a small amount of vacuum may be needed to get the flow started.**
- 11.2.2.8. Adjust the vacuum (or use no vacuum) to achieve a flow-rate of ~1 mL/min (~1 drop).
- 11.2.2.9. Discard column rinses.
- 11.2.2.10. Load sample solution slowly to each column at ~1 mL/min.  
**NOTE: It is likely that the ~1 mL/min, flow rate can be achieved with no vacuum at all. The frit should be pressed down tightly to prevent too fast a flow rate.**
- 11.2.2.11. Add 5mL of 1.5M HCl to rinse each sample solution tube and add to column at ~1–2 mL/min. Discard eluate.
- 11.2.2.12. Press frit down on resin bed.
- 11.2.2.13. Add 30 mL of 3M HCl to each column at ~1–2 mL/min. Discard rinse.  
**NOTE: The flow rate should not be too fast to ensure effective removal of Ca and other interferences.**
- 11.2.2.14. Press frit down tightly on resin bed.
- 11.2.2.15. Place clean 50 mL centrifuge tubes beneath the columns to catch the eluate.
- 11.2.2.16. Press frit down tightly on resin bed.
- 11.2.2.17. Add 20 mL of 8M of HNO<sub>3</sub> to each column to elute Ra at ~1mL/min. Record the date and time as the date and time of separation of <sup>225</sup>Ra and thorium to account for the decay of unsupported <sup>225</sup>Ra.  
**NOTE: Date and time need only be recorded if the <sup>225</sup>Ra was in equilibrium with <sup>229</sup>Th tracer.**
- 11.2.2.18. Transfer the eluate solution to 150 mL glass beakers. Rinse tubes with ~3 mL of 8M HNO<sub>3</sub> and add to beaker.

11.2.2.19. Add 2 mL of 30 wt% H<sub>2</sub>O<sub>2</sub> to each beaker and evaporate on medium heat to dryness on a hotplate being very careful not to bake material into the beaker.

11.2.2.20. Add 5 mL of 4M HNO<sub>3</sub> to redissolve each sample, warming slightly on hotplate as needed.

**NOTE: Barium in the sample can interfere with the <sup>226</sup>Ra alpha peak resolution. Sr Resin is used to remove Ba in the sample. The volume of 4M HNO<sub>3</sub> must be kept small to remove Ba effectively. Additional interferences (Ca, U, Th) are removed using DGA Resin.**

### 11.2.3. Separation of Barium and Radionuclide Interferences

11.2.3.1. Place a stacked column with 2 mL Sr Resin cartridge (top) and 2 mL DGA resin cartridge (bottom) on the vacuum box.

11.2.3.2. Condition each column with 5 mL of 4M HNO<sub>3</sub> at 1 mL/min. Discard rinse.

11.2.3.3. Ensure that clean, labeled plastic tubes are placed in the tube rack under each cartridge.

11.2.3.4. Transfer each sample solution from Step 11.2.2.20 into the appropriate column at a flow rate of ~1 mL/min or less.

11.2.3.5. Add 3 mL of 4M HNO<sub>3</sub> to each beaker (from Step 11.2.2.20) as a rinse and transfer each solution into the appropriate column at ~1 mL/min.

11.2.3.6. Add 5 mL of 4M HNO<sub>3</sub> into each reservoir as a column rinse (flow rate ~1-2 mL/min).

11.2.3.7. Record the date and time of the last rinse (Step 11.2.3.6) as the date and time of separation of radium from progeny. This is also the beginning of ingrowth of <sup>225</sup>Ac (and <sup>221</sup>Fr and <sup>217</sup>At).

**NOTE: If purified <sup>225</sup>Ra tracer is added to the sample (see Appendix that follows this method), the <sup>225</sup>Ra activity was unsupported before the tracer solution was added to the sample. The activity reference date and time established during standardization of the <sup>225</sup>Ra tracer is used as the reference date for the <sup>225</sup>Ra solution.**

**NOTE: If <sup>225</sup>Ra at some degree of secular equilibrium with <sup>229</sup>Th is added as tracer in the initial step, the activity of <sup>225</sup>Ra is dependent upon the total amount of time between the last <sup>229</sup>Th purification and cation exchange elution step (Step 11.2.2.17). The decay of <sup>225</sup>Ra starts at the <sup>229</sup>Th removal step and is decayed to the DGA Resin separation time, where <sup>225</sup>Ac is removed, to determine the reference activity of the <sup>225</sup>Ra tracer at that point.**

11.2.3.8. Turn off vacuum. Discard Sr Resin plus DGA Resin.

11.2.3.9. Remove tubes and transfer sample solution to 250 mL glass beakers, rinsing tube with 3 mL of 30 wt% H<sub>2</sub>O<sub>2</sub> and adding to beaker.

11.2.3.10. Evaporate solutions to dryness on a hotplate using medium heat. Do not bake.

11.2.3.11. Add 10 ml 1.5M HCl to each beaker to redissolve each sample, warming on a hotplate until hot and transfer to a labeled 50 mL tube.

11.2.3.12. Rinse each beaker with two 7 mL volumes of 1.5M HCl and transfer each rinse to the tube.

11.3. Barium sulfate micro-precipitation of  $^{226}\text{Ra}$

11.3.1. Add ~3.0 g of  $(\text{NH}_4)_2\text{SO}_4$  to the purified sample solution. Mix well using a vortex stirrer to completely dissolve the salt.

11.3.2. Add 50  $\mu\text{g}$  of Ba carrier (50  $\mu\text{L}$  of 1000  $\mu\text{g}$  Ba/mL) into each tube. Cap and mix well with vortex stirrer.

11.3.3. Add 5.0 mL of isopropanol and mix well using a vortex stirrer.

11.3.4. Place each tube in an ice bath filled with cold tap water for at least 15 minutes, periodically stirring on vortex stirrer (before placing in ice, midway, and after icing).

11.3.5. Pre-wet a 0.1-micron filter using methanol or ethanol. Filter the suspension through the filter using vacuum. The precipitate *will not be* visually apparent.

11.3.6. Rinse the sample container with 3 mL of 20% isopropanol solution.

11.3.7. Rinse the filter apparatus with about 2 mL of methanol or ethanol to facilitate drying. Turn off vacuum and discard rinses.

11.3.8. Mount the filter on a labeled adhesive mounting disk (or equivalent) ensuring that the filter is not wrinkled and is centered on mounting disk.

11.3.9. Place the filter under a heat lamp for approximately 5 minutes or more until it is completely dry.

11.3.10. Store the filter for approximately 24 hours to allow sufficient  $^{217}\text{At}$  (third progeny of  $^{225}\text{Ra}$ ) to ingrow into the sample test source allowing a measurement uncertainty for the  $^{217}\text{At}$  of  $< 5\%$ .

**NOTE: The filter may be counted immediately if a long enough count time is used to allow enough tracer counts to accumulate while the sample is being counted. For example, a longer 1000 minute may be used so the ingrowth of  $^{217}\text{At}$  and sample counting can be done simultaneously. This reduces the overall analytical time required.**

11.3.11. Count by alpha spectrometry. The count times should be adjusted to meet the uncertainties and detection capabilities identified in Steps 9.3, 9.4, and 9.5.

12. Data Analysis and Calculations

12.1. The final sample test source (filter mounted on a planchet) will likely need to have approximate ingrowth period of 18 to 24 hours for  $^{225}\text{Ac}$  (and  $^{221}\text{Fr}$  and  $^{217}\text{At}$ ) to meet Analytical Protocol Specifications for chemical yield with a counting time of 4 to 8 hours. At-217 (third progeny of  $^{225}\text{Ra}$ ) has a single, distinct alpha peak with a centroid at 7.067 MeV and is used for determining the yield.

12.2. The following equation can be used to calculate the radiochemical yield:

$$RY = \frac{R_t - R_b}{\varepsilon \times A_t \times I_t} \quad (1)$$

where:

- $RY$  = Fractional radiochemical yield based on  $^{225}\text{Ra}$  (from ingrown  $^{217}\text{At}$  at 7.07 MeV)  
 $R_t$  = Total count rate beneath the  $^{217}\text{At}$  peak at 7.07 MeV, cpm  
 $R_b$  = Background count rate for the same region, cpm  
 $\varepsilon$  = Efficiency for the alpha spectrometer  
 $I_t$  = Fractional abundance for the 7.07 MeV alpha peak counted (= 0.9999)

**NOTE: If  $^{225}\text{Ra}$  is separated from  $^{229}\text{Th}$  for use as a purified tracer, the  $^{225}\text{Ra}$  activity is unsupported and begins to decay at time of prior separation from  $^{229}\text{Th}$ . The reference date and time established when the tracer is standardized is used for decay correction of the  $^{225}\text{Ra}$  activity. If  $^{229}\text{Th}$  solution (with  $^{225}\text{Ra}$  in full secular equilibrium) is added to the sample, the  $^{225}\text{Ra}$  activity is equal to the  $^{229}\text{Th}$  activity added and only begins to decay at the point of separation of  $^{225}\text{Ra}$  from  $^{229}\text{Th}$  during the sample preconcentration steps (cation exchange elution step).**

$$A_t = \text{Activity of } ^{217}\text{At} \text{ at midpoint of the count (the target value that should be achieved for 100\% yield), in dpm}$$

$$= 3.0408(I_t)(A_{225\text{Ra}}) [e^{-\lambda_1 d} - e^{-\lambda_2 d}] \quad (2)$$

$A_{225\text{Ra}}$  = Activity in dpm of  $^{225}\text{Ra}$  tracer added to the sample decay corrected to the date and time of radium separation in Step 11.3.6.<sup>2</sup>

$d$  = Elapsed ingrowth time for  $^{225}\text{Ac}$  [and the progeny  $^{217}\text{At}$ ], in days from the date and time of Ra separation to the midpoint of the sample count

$\lambda_1$  = 0.04652 d<sup>-1</sup> (decay constant for  $^{225}\text{Ra}$  – half-life = 14.9 days)

$\lambda_2$  = 0.06931 d<sup>-1</sup> (decay constant for  $^{225}\text{Ac}$ ) – half-life = 10.0 days)

<sup>2</sup> **Unsupported  $^{225}\text{Ra}$ :** When separated  $^{225}\text{Ra}$  tracer is added to the sample, its initial activity,  $A_{225\text{Ra-initial}}$ , must be corrected for decay from the reference date established during standardization of the tracer to the point of separation of  $^{225}\text{Ra}$  and  $^{225}\text{Ac}$  as follows:

$$A_{225\text{Ra}} = (A_{225\text{Ra-initial}})(e^{-\lambda_1 d_i})$$

where:  $\lambda_1$  = decay constant for  $^{225}\text{Ra}$  (0.04652 d<sup>-1</sup>); and  $d_i$  = time elapsed between the activity reference date for the  $^{225}\text{Ra}$  tracer solution added to the sample and the separation of  $^{225}\text{Ra}$  and  $^{225}\text{Ac}$  in Step 11.2.3.6 (days).

**$^{229}\text{Th}/^{225}\text{Ra}$  added in equilibrium:** When  $^{229}\text{Th}$  containing ingrown  $^{225}\text{Ra}$  is added directly to the sample, the amount of  $^{225}\text{Ra}$  ingrown since purification of the  $^{229}\text{Th}$  solution up until  $^{229}\text{Th}$  removal point during the method is calculated as:

$$A_{225\text{Ra}} = (A_{229\text{Th}})(1 - e^{-\lambda_1 d_i})$$

where:  $A_{229\text{Th}}$  = Activity of the  $^{229}\text{Th}$  standard on the date of the separation of Th and Ra (cation exchange elution step);  $\lambda_1$  = decay constant for  $^{225}\text{Ra}$  (0.04652 d<sup>-1</sup>); and  $d_i$  = time elapsed between the purification of  $^{229}\text{Th}$  solution added to the sample and the separation of  $^{225}\text{Ra}$  and  $^{229}\text{Th}$  (days). The  $^{225}\text{Ra}$  is then corrected for decay to the  $^{225}\text{Ac}$  removal separation time (Step 11.2.3.6) using the first equation above.



$I_t$  = Fractional abundance for the 7.07 MeV alpha peak counted (= 0.9999)  
 3.0408 =  $\lambda_2/(\lambda_2 + \lambda_1)$  [a good approximation as the half lives of  $^{221}\text{Fr}$  and  $^{217}\text{At}$  are short enough so that secular equilibrium with  $^{225}\text{Ac}$  is ensured]

12.3. The activity concentration of an analyte and its combined standard uncertainty are calculated using the following equations:

$$AC_a = \frac{A_t \times R_{na}}{W_a \times R_{nt} \times D_a \times I_a \times 2.22} \quad (3)$$

and

$$u_c(AC_a) = \sqrt{u^2(R_{na}) \times \frac{A_t^2}{W_a^2 \times R_{nt}^2 \times D_a^2 \times I_a^2 \times 2.22^2} + AC_a^2 \times \left( \frac{u^2(A_t)}{A_t^2} + \frac{u^2(W_a)}{W_a^2} + \frac{u^2(R_{nt})}{R_{nt}^2} \right)} \quad (4)$$

where:

- $AC_a$  = activity concentration of the analyte at time of count, (pCi/g)
- $A_t$  = activity of  $^{217}\text{At}$  at midpoint of the count (the target value that should be achieved for 100% yield), in dpm (See Step 12.2 for detailed calculation of  $A_t$ )
- $R_{na}$  = net count rate of the analyte in the defined region of interest (ROI), in counts per minute (*note that the peaks at 4.784 and 4.602 MeV are generally included in the ROI for  $^{226}\text{Ra}$* )
- $R_{nt}$  = net count rate of the tracer in the defined ROI, in counts per minute
- $W_a$  = weight of the sample aliquant (g)
- $D_a$  = correction factor for decay of the analyte from the time of sample collection (or other reference time) to the midpoint of the counting period, if required
- $I_a$  = probability of  $\alpha$  emission for  $^{226}\text{Ra}$  (the combined peaks at 4.78 and 4.602 MeV are generally included in the ROI with an abundance of 1.00.)<sup>3</sup>
- $u_c(AC_a)$  = combined standard uncertainty of the activity concentration of the analyte (pCi/g)
- $u(A_t)$  = standard uncertainty of the activity of the tracer added to the sample (dpm)
- $u(W_a)$  = standard uncertainty of the volume of sample aliquant (g)
- $u(R_{na})$  = standard uncertainty of the net count rate of the analyte in counts per minute
- $u(R_{nt})$  = standard uncertainty of the net count rate of the tracer in counts per minute

**NOTE: The uncertainties of the decay-correction factors and of the probability of decay factors are assumed to be negligible.**

<sup>3</sup> If the individual peak at 4.78 MeV used, and completely resolved from the 4.602 MeV peak, the abundance would be 0.9445.

**NOTE: The equation for the combined standard uncertainty ( $u_c(AC_a)$ ) calculation is arranged to eliminate the possibility of dividing by zero if  $R_a = 0$ .**

**NOTE: The standard uncertainty of the activity of the tracer added to the sample must reflect that associated with the activity of the standard reference material and any other significant sources of uncertainty such as those introduced during the preparation of the tracer solution (e.g., weighing or dilution factors) and during the process of adding the tracer to the sample.**

12.3.1. The net count rate of an analyte or tracer and its standard uncertainty can be calculated using the following equations:

$$R_{nx} = \frac{C_x}{t_s} - \frac{C_{bx}}{t_b} \quad (5)$$

and

$$u(R_{nx}) = \sqrt{\frac{C_x + 1}{t_s^2} + \frac{C_{bx} + 1}{t_b^2}} \quad (6)$$

where:

- $R_{nx}$  = net count rate of analyte or tracer, in counts per minute<sup>4</sup>
- $C_x$  = sample counts in the analyte or the tracer ROI
- $t_s$  = sample count time (min)
- $C_{bx}$  = background counts in the same ROI as for x (x refers to the respective analyte or tracer count)
- $t_b$  = background count time (min)
- $u(R_{nx})$  = standard uncertainty of the net count rate of tracer or analyte, in counts per minute

12.3.2. If the critical level concentration ( $L_c$ ) or the minimum detectable concentration (MDC) are requested (at an error rate of 5%), they can be calculated using the following equations.<sup>5</sup>

$$L_c = \frac{\left[ 0.4 \times \left( \frac{t_s}{t_b} - 1 \right) + 0.677 \times \left( 1 + \frac{t_s}{t_b} \right) + 1.645 \times \sqrt{\left( R_{ba} t_b + 0.4 \right) \times \frac{t_s}{t_b} \times \left( 1 + \frac{t_s}{t_b} \right)} \right] \times A_t \times D_a \times I_a}{t_s \times W_a \times R_t \times D_a \times I_a}$$

<sup>4</sup> For methods with very low counts, MARLAP Section 19.5.2.2 recommends adding one count each to the gross counts and the background counts when estimating the uncertainty of the respective net counts. This minimizes negative bias in the estimate of uncertainty and protects against calculating zero uncertainty when a total of zero counts are observed for the sample and background.

<sup>5</sup> The formulations for the critical level and minimum detectable concentration are based on the Stapleton Approximation as recommended in MARLAP Section 20A.2.2, Equations 20.54 and 20A.3.2, and Equation 20.74, respectively. The formulations presented here assume an error rate of  $\alpha = 0.05$ ,  $\beta = 0.05$  (with  $z_{1-\alpha} = z_{1-\beta} = 1.645$ ), and  $d = 0.4$ . For methods with very low numbers of counts, these expressions provide better estimates than do the traditional formulas for the critical level and MDC.

$$\text{MDC} = \frac{\left[ 2.71 \times \left( 1 + \frac{t_s}{t_b} \right) + 3.29 \times \sqrt{R_{ba} t_s \times \left( 1 + \frac{t_s}{t_b} \right)} \right] \times A_t}{t_s \times W_a \times R_{nt} \times D_a \times I_a \times 2.22} \quad (7)$$

where:

$R_{ba}$  = background count rate for the analyte in the defined ROI, in counts per minute

#### 12.4. Results Reporting

12.4.1. The following data should be reported for each result: weight of sample used; yield of tracer and its uncertainty; and full width at half maximum of each peak used in the analysis.

12.4.2. The following conventions should be used for each result:

12.4.2.1. Result in scientific notation  $\pm$  combined standard uncertainty.

#### 13. Method Performance

13.1. Results of method validation performance are to be archived and available for reporting purposes.

13.2. Expected sample preparation time for a batch of 15 samples is approximately seven hours. Total processing time is dependent on actual wait time for  $^{217}\text{At}$  ingrowth (approximately 16–24 hours) and count times (approximately 6 hours).

#### 14. Pollution Prevention

14.1. The use of 50WX8 cation resin, Sr Resin and DGA Resin reduces the amount of solvents that would otherwise be needed to co-precipitate and purify the final sample test source.

#### 15. Waste Management

15.1. Nitric acid and hydrochloric acid wastes should be neutralized before disposal and then disposed of in accordance with local ordinances.

15.2. All final precipitated materials contain tracer and should be dealt with as radioactive waste and disposed of in accordance with the restrictions provided in the facility's NRC and/or state license.

15.3. It may be advisable to rinse the cation resin columns with water to remove strong nitric acid prior to resin disposal.

#### 16. References

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1 17. Tables, Diagrams, and Flow Charts  
 2 17.1. Tables [including major radiation emissions from all radionuclides separated]  
 3  
 4

**Table 17.1 – Alpha Particle Energies and Abundances of Importance**

Energy (MeV)	Abundance (%)	Nuclide	Energy (MeV)	Abundance (%)	Nuclide
4.601	5.6	Ra -226	5.791	8.6	Ac -225
4.784	94.5	Ra -226	5.793	18.1	Ac -225
4.798	1.5	Th -229	5.830	50.7	Ac -225
4.815	9.3	Th -229	5.869	1.9	Bi -213
4.838	5.0	Th -229	6.002	100.0	Po -218
4.845	56.2	Th -229	6.051	25.1	Bi -212
4.901	10.2	Th -229	6.090	9.8	Bi -212
4.968	6.0	Th -229	6.126	15.1	Fr -221
4.979	3.2	Th -229	6.243	1.3	Fr -221
5.053	6.6	Th -229	6.278	16.2	Bi -211
5.434	2.2	Ra -223	6.288	99.9	Rn -220
5.449	5.1	Ra -224	6.341	83.4	Fr -221
5.489	99.9	Rn -222	6.425	7.5	Rn -219
5.540	9.0	Ra -223	6.553	12.9	Rn -219
5.580	1.2	Ac -225	6.623	83.5	Bi -211
5.607	25.2	Ra -223	6.778	100.0	Po -216
5.609	1.1	Ac -225	6.819	79.4	Rn -219
5.637	4.4	Ac -225	7.067	99.9	Po -217
5.682	1.3	Ac -225	7.386	100.0	Po -215
5.685	94.9	Ra -224	7.450	98.9	Po -211
5.716	51.6	Ra -223	7.687	100.0	Po -214
5.724	3.1	Ac -225	8.376	100.0	Po -213
5.732	8.0	Ac -225	8.525	2.1	Po -212
5.732	1.3	Ac -225	11.660	96.8	Po -212
5.747	9.0	Ra -223			

 - Analyte      - <sup>217</sup>At (3rd progeny of <sup>225</sup>Ra tracer)  
 - <sup>229</sup>Th (Check ROI for indications of inadequate clean-up)

5  
 6 *Includes only alpha particles with abundance > 1%.*  
 7 *Reference: NUDAT 2.4, Radiation Decay National Nuclear Data Center, Brookhaven National Laboratory;*  
 8 *Available at: [www.nndc.bnl.gov/nudat2/indx\\_dec.jsp](http://www.nndc.bnl.gov/nudat2/indx_dec.jsp); Queried: November 11, 2007.*  
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**Table 17.2 – Ingrowth Factors for <sup>217</sup>At in <sup>225</sup>Ra**

<b>Time elapsed between separation of Ra and midpoint of count in hours</b>	<b>1</b>	<b>2</b>	<b>3</b>	<b>4</b>	<b>5</b>	<b>6</b>	<b>24</b>	<b>48</b>
<b>Ingrowth Factor*</b>	0.002881	0.005748	0.008602	0.01144	0.01427	0.01708	0.06542	0.1235
<b>Time elapsed between separation of Ra and midpoint of count in hours</b>	<b>72</b>	<b>96</b>	<b>120</b>	<b>144</b>	<b>192</b>	<b>240</b>	<b>360</b>	<b>480</b>
<b>Ingrowth Factor*</b>	0.1748	0.2200	0.2596	0.2940	0.3494	0.3893	0.4383	0.4391

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\*Ingrowth Factor represents the fraction of <sup>217</sup>Ac activity at the midpoint of the sample count relative to the <sup>225</sup>Ra activity present at the date/time of Ra separation. These ingrowth factors may be closely approximated (within a fraction of a percent) using the expression for A<sub>1</sub> in Step 12.2.

**Table 17.3 – Ingrowth Factors for <sup>225</sup>Ra in <sup>229</sup>Th**

<b>Time elapsed between purification of the <sup>229</sup>Th standard and date of Ra separation in days</b>	<b>1</b>	<b>5</b>	<b>10</b>	<b>12</b>	<b>15</b>	<b>20</b>	<b>25</b>	<b>27</b>	<b>30</b>	<b>40</b>
<b>Ingrowth Factor*</b>	0.04545	0.2075	0.3720	0.4278	0.5023	0.6056	0.6875	0.7152	0.7523	0.8445
<b>Time elapsed between purification of the <sup>229</sup>Th standard and date of Ra separation in days</b>	<b>50</b>	<b>55</b>	<b>60</b>	<b>70</b>	<b>80</b>	<b>90</b>	<b>100</b>	<b>130</b>	<b>160</b>	<b>200</b>
<b>Ingrowth Factor*</b>	0.9023	0.9226	0.9387	0.9615	0.9758	0.9848	0.9905	0.9976	0.9994	0.9999

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\*Ingrowth Factor represents the fraction <sup>225</sup>Ra activity/<sup>229</sup>Th activity at the time of Ra separation.

**Table 17.4 Decay Factors for Unsupported <sup>225</sup>Ra**

<b>Time elapsed between separation of <sup>229</sup>Th and <sup>225</sup>Ra in days</b>	<b>1</b>	<b>5</b>	<b>10</b>	<b>12</b>	<b>15</b>	<b>20</b>	<b>25</b>	<b>27</b>	<b>30</b>	<b>40</b>
<b>Decay Factor*</b>	0.9545	0.7925	0.6280	0.5722	0.4977	0.3944	0.3125	0.2848	0.2477	0.1555
<b>Time elapsed between separation of <sup>229</sup>Th and <sup>225</sup>Ra in days</b>	<b>50</b>	<b>55</b>	<b>60</b>	<b>70</b>	<b>80</b>	<b>90</b>	<b>100</b>	<b>130</b>	<b>160</b>	<b>200</b>
<b>Decay Factor*</b>	0.09769	0.07741	0.06135	0.03853	0.02420	0.01519	0.00954	0.00236	0.00059	0.00009

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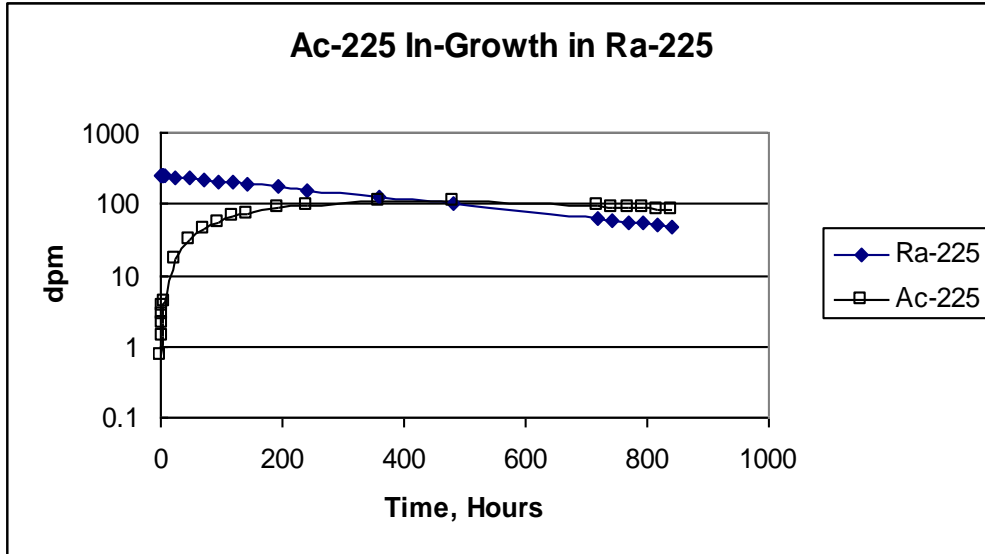
20

21

\*Decay Factor represents the fraction <sup>225</sup>Ra activity remaining as calculated using the equation in Footnote 2.

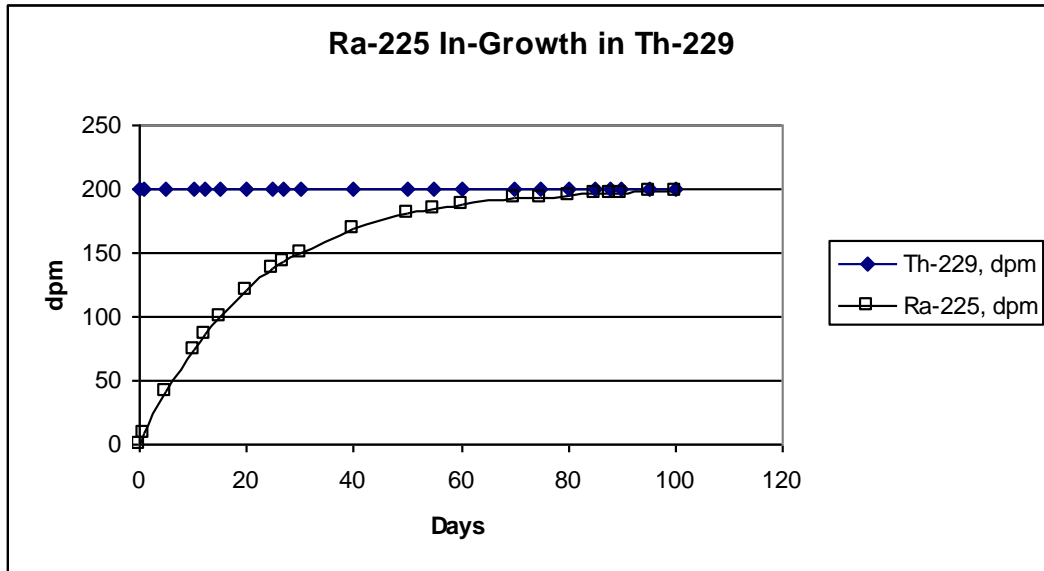
22 17.2. Ingrowth curves and Ingrown factors

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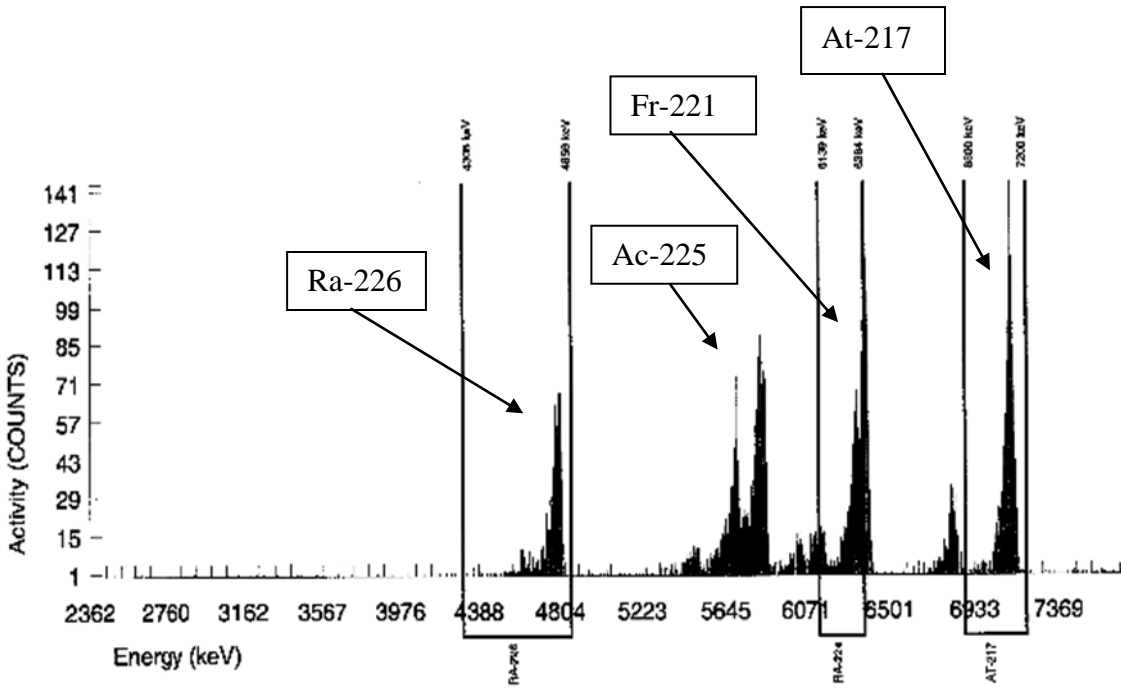
25



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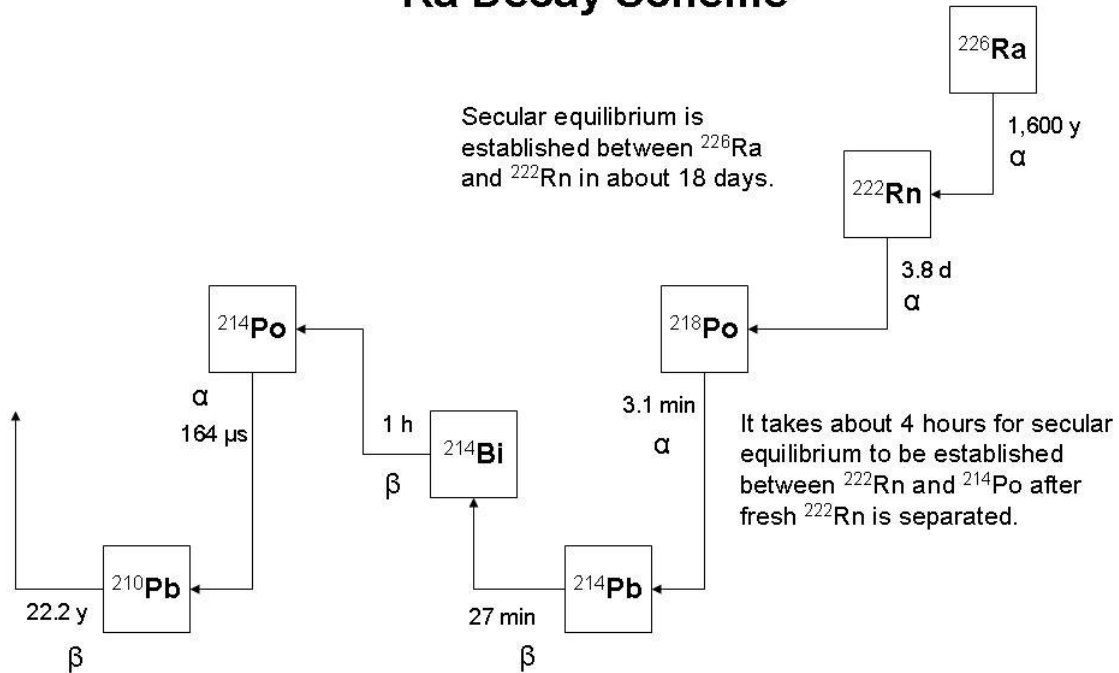
27

28 17.3. Example Alpha Spectrum from a Processed Sample



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30  
31 17.4. Decay Schemes for Analyte and Tracer

**<sup>226</sup>Ra Decay Scheme**

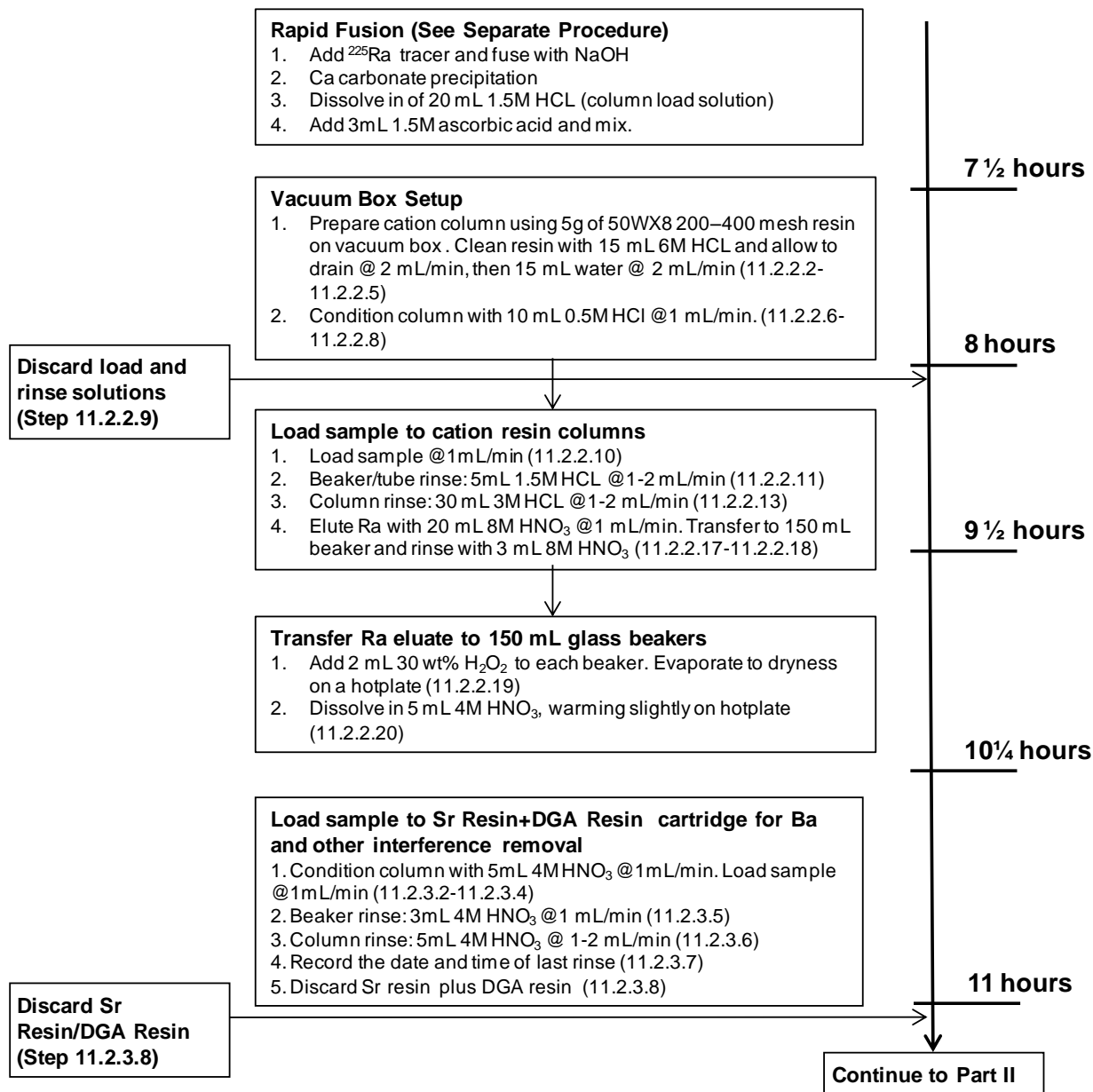




32 17.5 Flow Charts: Separation Scheme and Timeline for Determination of Ra-226 in  
 33 Building Materials (Parts 1 and 2)

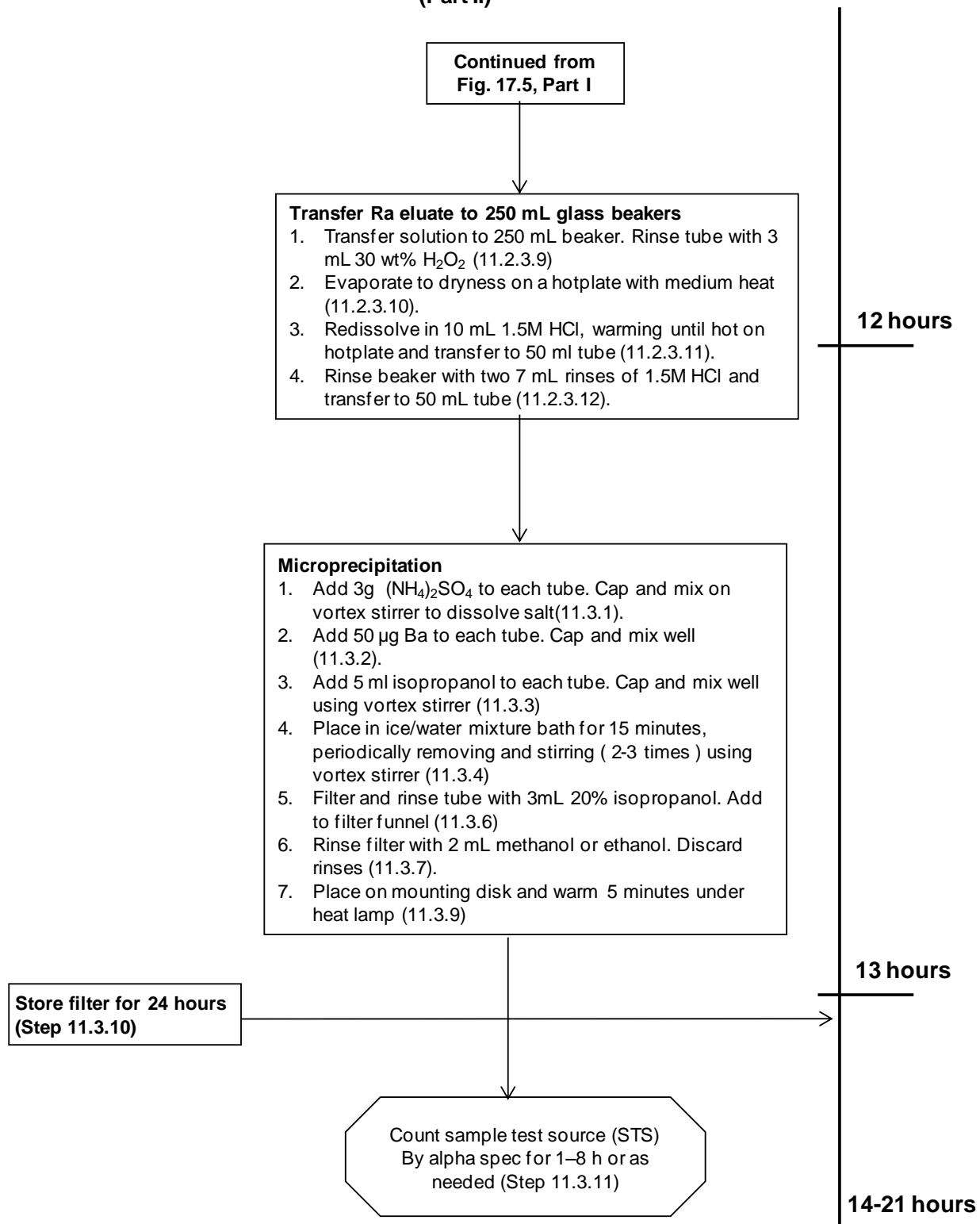
34 **Note: The example timeline below illustrates the sample preparation for asphalt shingles, which**  
 35 **requires the destruction of organics. The sample preparation times will be shorter when furnace**  
 36 **ashing steps to destroy organics prior to the fusion are not required.**

**Separation Scheme and Timeline for Determination of  
 Ra-226 in Building Materials  
 (Part I)**



37

**Separation Scheme and Timeline for Determination of Ra-226 in Building Materials (Part II)**



## **Appendix A: Preparation and Standardization of $^{225}\text{Ra}$ Tracer Following Separation from $^{229}\text{Th}$**

### A1. Summary Description of Procedure

This procedure describes a  $^{225}\text{Ra}$  generator to make tracer amounts of  $^{225}\text{Ra}$  using a  $^{229}\text{Th}$  solution. Thorium-229 is separated from  $^{225}\text{Ra}$  using yttrium hydroxide ( $\text{Y}(\text{OH})_3$ ) co-precipitation. Thorium-229 is carried in the precipitate and most of the  $^{225}\text{Ra}$  remains in solution. Centrifugation to remove  $^{229}\text{Th}$  in the precipitate and filtration of the supernate produces the  $^{225}\text{Ra}$  tracer solution. The  $^{225}\text{Ra}$  activity of the tracer solution is standardized by counting sample test sources prepared from at least five replicate aliquants of the  $^{225}\text{Ra}$  solution, each spiked with a known quantity of a  $^{226}\text{Ra}$  standard. This standardized activity concentration, referenced to the date and time of the  $^{225}\text{Ra}$  separation described in Step A4.10.7, is then decay-corrected to the date and time of subsequent sample analyses.

The  $\text{Y}[\text{Th}](\text{OH})_3$  precipitate may be stored and re-used later to generate more  $^{225}\text{Ra}$  tracer solution. Radium-225 ingrows in the  $^{229}\text{Th}$  fraction ( $\text{Y}(\text{OH})_3$  precipitate) and after 50 days will be about 90% ingrown. After sufficient ingrowth time  $^{225}\text{Ra}$  may be harvested to make a fresh  $^{225}\text{Ra}$  tracer solution by dissolving the precipitate and re-precipitating  $\text{Y}(\text{OH})_3$  to separate  $^{229}\text{Th}$  from  $^{225}\text{Ra}$ .

Multiple  $^{225}\text{Ra}$  generators may be prepared to ensure that  $^{225}\text{Ra}$  tracer will be continuously available. The  $^{225}\text{Ra}$  tracer solution produced is usable for 2–3 half-lives (approximately 30–45 days). To minimize effort involved with standardization of the  $^{225}\text{Ra}$  solution, it is recommended that the laboratory prepare an amount of  $^{229}\text{Th}$  sufficient to support the laboratory's expected workload for 3–5 weeks. Since the  $^{229}\text{Th}$  solution is reused, and the half-life of  $^{229}\text{Th}$  is long (7,342 years), the need to purchase a new certified  $^{229}\text{Th}$  solution is kept to a minimum.

### A2. Equipment and Supplies

A2.1. Refer to Section 6 of the main procedure.

### A3. Reagents and Standards

A3.1. Refer to Section 7 of the main procedure.

### A4. Procedure

- A4.1. Add a sufficient amount of  $^{229}\text{Th}$  solution (that which will yield at least 150–600 dpm/mL of the  $^{225}\text{Ra}$  solution) to a 50 mL centrifuge tube.<sup>6</sup>
- A4.2. Add 20 mg yttrium (Y) (2 mL of 10 mg/mL Y metals standard stock solution).
- A4.3. Add 1 mg Ba (0.1 mL of 10 mg/mL Ba metals standard stock solution).
- A4.4. Add 4 mL of concentrated ammonium hydroxide to form  $\text{Y}(\text{OH})_3$  precipitate.
- A4.5. Centrifuge and decant the supernatant into the open barrel of a 50 mL syringe, fitted with a 0.45  $\mu\text{m}$  syringe filter. Hold the syringe barrel over a new 50 mL centrifuge

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<sup>6</sup> For example, if 40 mL of a  $^{229}\text{Th}$  solution of 600 dpm/mL is used, the maximum final activity of  $^{225}\text{Ra}$  will be ~510 dpm/mL at Step A4.7. This solution would require about 1.4 mL for the standardization process and about 8 mL for a batch of 20 samples.

- tube while decanting. Insert the syringe plunger and filter the supernatant into the new centrifuge tube. Discard the filter as potentially contaminated radioactive waste.
- A4.6. Cap the centrifuge tube with the precipitate, label clearly with the standard ID, precipitation date, and the technician's initials and store for future use.
- A4.7. Properly label the new centrifuge tube with the supernate. This is the  $^{225}\text{Ra}$  tracer solution.
- A4.8. Add 3 mL of concentrated HCl to  $^{225}\text{Ra}$  tracer solution. Cap centrifuge tube and mix well.
- A4.9. Prepare the following solutions in 10 mL of 4M  $\text{HNO}_3$  for standardization of  $^{225}\text{Ra}$  tracer.

<b><u>Solution</u></b>	<b><u>Spike(s)</u></b>
Standardization Replicates (5 replicates)	~80 dpm of the $^{225}\text{Ra}$ tracer solution, and ~8 dpm of a $^{226}\text{Ra}$ standard traceable to the National Institute of Standards and Technology (NIST) or equivalent
Blank	~80 dpm of the $^{225}\text{Ra}$ tracer solution ( <i>the blank should be evaluated to confirm that <math>^{226}\text{Ra}</math> is not detected in the <math>^{225}\text{Ra}</math> tracer solution at levels that may compromise sample results when used in the method</i> )
Standardization Control Sample	~80 dpm of the $^{225}\text{Ra}$ tracer solution, and ~8 dpm of a second source independent traceable $^{226}\text{Ra}$ standard (the Standardization Control Sample should be evaluated to confirm that the standardiza- tion process does not introduce significant bias into the standardized value for the $^{225}\text{Ra}$ tracer).

- A4.10. Process the solutions to prepare sources for alpha spectrometry as follows:
- A4.10.1. Place  $^{225}\text{Ra}$  aliquots of standard (A4.9) in 10 mL of 4M  $\text{HNO}_3$  in 50 mL glass beakers.
- A4.10.2. Place a 2 mL DGA Resin cartridge on the vacuum box.
- A4.10.3. Add 5 mL of 4M  $\text{HNO}_3$  into each column to precondition resin at ~1 mL/min. Discard rinse.
- A4.10.4. Transfer each sample solution from Step A4.10.1 into the appropriate column reservoir. Allow solution to pass through the DGA Resin cartridge at a flow rate of ~1 mL/min or less.
- A4.10.5. Add 5 mL of 4M  $\text{HNO}_3$  to each beaker (from Step A4.10.1) as a rinse and transfer each solution into the appropriate reservoir at ~1 mL/min.
- A4.10.6. Add 5 mL of 4M  $\text{HNO}_3$  into each column to rinse at ~1 mL/min.
- A4.10.7. Record the date and time of the last rinse as the date and time of separation of radium (beginning of  $^{225}\text{Ac}$  ingrowth).

**NOTE: The activity reference date and time established during standardization of the  $^{225}\text{Ra}$  tracer is used as the reference date for the  $^{225}\text{Ra}$  solution.**

- A4.10.8. Remove tubes from vacuum box and evaporate to dryness.
- A4.10.9. Remove tubes and transfer sample solution to 250 mL glass beakers, rinsing tube with 3 mL of 30 wt%  $\text{H}_2\text{O}_2$  and adding to beaker.
- A4.10.10. Evaporate solutions to dryness on a hotplate using medium heat. Do not bake.
- A4.10.11. Add 10 ml 1.5M HCl to each beaker to redissolve each sample, warming on a hotplate until hot and transfer to a labeled 50 mL tube.
- A4.10.12. Rinse each beaker with two 7 mL volumes of 1.5M HCl and transfer each rinse to the tube.
- A4.10.13. Add ~3.0 g of  $(\text{NH}_4)_2\text{SO}_4$  to the purified sample solution Mix well to completely dissolve the salt (dissolves readily).
- A4.10.14. Add 75  $\mu\text{g}$  of Ba carrier (75  $\mu\text{L}$  of 1000  $\mu\text{g}$  Ba/mL) into each tube. Cap and mix well with vortex stirrer.
- A4.10.15. Add 5.0 mL of isopropanol and mix well using a vortex stirrer.
- A4.10.16. Place each tube in an ice bath filled with cold tap water for at least 20 minutes, periodically stirring on vortex stirrer.  
**NOTE: Sonication may be used instead of occasional stirring using a vortex stirrer.**
- A4.10.17. Pre-wet a 0.1-micron filter using methanol or ethanol. Filter the suspension through the filter using vacuum. The precipitate *will not be* visually apparent.
- A4.10.18. Rinse the sample container with 3 mL of 20% isopropanol solution.
- A4.10.19. Rinse the filter apparatus with about 2 mL of methanol or ethanol to facilitate drying. Turn off vacuum.
- A4.10.20. Mount the filter on a labeled adhesive mounting disk (or equivalent) ensuring that the filter is not wrinkled and is centered on mounting disk.
- A4.10.21. Place the filter under a heat lamp for ~5 minutes or more until it is completely dry.
- A4.10.22. Count filters for an appropriate period of time by alpha spectrometry.
- A4.10.23. Mount the dried filter on a support appropriate for the counting system to be used.
- A4.10.24. Store the filter for at least 24 hours to allow sufficient  $^{217}\text{At}$  (third progeny of  $^{225}\text{Ra}$ ) to ingrow into the sample test source allowing a measurement uncertainty for the  $^{217}\text{At}$  of < 5 %.
- A4.10.25. After allowing about 24-hours ingrowth, count the standardization sources by alpha spectrometry.
- A4.11. Calculate the activity of  $^{225}\text{Ra}$ , in units of dpm/mL, in the standardization replicates, at the  $^{225}\text{Ra}$  time of separation as follows:

$$AC_{225Ra} = \frac{\left( \frac{N_{217At}}{t_{217At}} - \frac{N_b}{t_b} \right) \times (A_{226Ra}) \times (V_{226Ra})}{\left( \frac{N_{226Ra}}{t_a} - \frac{N_b}{t_b} \right) \times \left[ (3.0408) (I_{217At}) (e^{-\lambda_1 d} - e^{-\lambda_2 d}) \right] \times V_{225Ra}} \quad (1)$$

where:

- $AC_{225Ra}$  = Activity concentration of  $^{225}\text{Ra}$ , in dpm/mL [at the time of separation from  $^{229}\text{Th}$ , Step A4.10.7]
- $N_{217At}$  = Total counts beneath the  $^{217}\text{At}$  peak at 7.07 MeV
- $N_{226Ra}$  = Total counts beneath the  $^{226}\text{Ra}$  peak at 4.78 MeV
- $N_b$  = Background count rate for the corresponding region of interest,
- $t_a$  = Duration of the count for the sample test source, minutes
- $t_b$  = Duration of the background count, minutes
- $A_{226Ra}$  = Activity of  $^{226}\text{Ra}$  added to each aliquant, in dpm/mL
- $V_{226Ra}$  = Volume of  $^{226}\text{Ra}$  solution taken for the analysis (mL)
- $V_{225Ra}$  = Volume of  $^{225}\text{Ra}$  solution taken for the analysis (mL)
- $d$  = Elapsed ingrowth time for  $^{225}\text{Ac}$  [and the progeny  $^{217}\text{At}$ ], from separation to the midpoint of the sample count, days
- $\lambda_1$  =  $0.04652 \text{ d}^{-1}$  (decay constant for  $^{225}\text{Ra}$  – half-life = 14.9 days)
- $\lambda_2$  =  $0.06931 \text{ d}^{-1}$  (decay constant for  $^{225}\text{Ac}$  – half-life = 10.0 days)
- $I_{217At}$  = Fractional abundance for the 7.07 MeV alpha peak counted (= 0.9999)
- 3.0408 =  $\lambda_2 / (\lambda_2 - \lambda_1)$  [a good approximation as the half lives of  $^{221}\text{Fr}$  and  $^{217}\text{At}$  are short enough so secular equilibrium with  $^{225}\text{Ac}$  is ensured]

**NOTE: The activity of the separated  $A_{225Ra}$  will need to be decay corrected to the point of separation in the main procedure (Step 11.2.3.6) so that the results can be accurately determined.**

A4.12. Calculate the uncertainty of the activity concentration of the  $^{225}\text{Ra}$  tracer at the reference date/time:

$$u_c(AC_{225Ra}) = \sqrt{\frac{\left( \frac{N_{217At}}{t_a} + \frac{N_b}{t_b} \right) \times AC_{226Ra}^2 \times I_{226Ra}^2 \times V_{226Ra}^2}{\left( \frac{N_{226Ra}}{t_a} - \frac{N_b}{t_b} \right)^2 \times [3.0408 \times I_{217At} \times (e^{-\lambda_1 d} - e^{-\lambda_2 d})]^2 \times V_{225Ra}^2} + AC_{225Ra}^2 \times \left( \frac{u^2(AC_{226Ra})}{AC_{226Ra}^2} + \frac{u^2(V_{225Ra})}{V_{225Ra}^2} + \frac{u^2(V_{226Ra})}{V_{226Ra}^2} + \frac{u^2(R_{226Ra})}{R_{226Ra}^2} \right)} \quad (2)$$

where:

- $u(AC_{225Ra})$  = Standard uncertainty of the activity concentration of  $^{225}\text{Ra}$ , in dpm/mL
- $N_{217At}$  = Total counts beneath the  $^{217}\text{At}$  peak at 7.07 MeV,
- $N_{226Ra}$  = Total counts beneath the  $^{226}\text{Ra}$  tracer peak at 4.78 MeV
- $N_b$  = Background count rate for the corresponding region of interest,
- $t_a$  = Duration of the count for the sample test source, minutes
- $t_b$  = Duration of the background count, minutes
- $AC_{226Ra}$  = Activity of  $^{226}\text{Ra}$  added to each aliquant, in dpm/mL

$u(AC_{226\text{Ra}})$	= Standard uncertainty of activity of $^{225}\text{Ra}$ , in dpm/mL
$V_{226\text{Ra}}$	= Volume of $^{226}\text{Ra}$ solution taken for the analysis (mL)
$u(V_{226\text{Ra}})$	= Standard uncertainty of the volume of $^{226}\text{Ra}$ solution taken for the analysis (mL)
$I_{226\text{Ra}}$	= Fractional abundance for the $^{226}\text{Ra}$ peak at 4.78 MeV (= 1.000)
$V_{225\text{Ra}}$	= Volume of $^{225}\text{Ra}$ solution taken for the analysis (mL)
$u(V_{225\text{Ra}})$	= Standard uncertainty of volume of $^{225}\text{Ra}$ solution taken for the analysis (mL)
$d$	= Elapsed ingrowth time for $^{225}\text{Ac}$ [and the progeny $^{217}\text{At}$ ], from separation to the midpoint of the sample count, days
$\lambda_1$	= $0.04652 \text{ d}^{-1}$ (decay constant for $^{225}\text{Ra}$ – half-life = 14.9 days)
$\lambda_2$	= $0.06931 \text{ d}^{-1}$ (decay constant for $^{225}\text{Ac}$ ) – half-life = 10.0 days)
3.0408	= $\lambda_2 d / (\lambda_2 d - \lambda_1 d)$ [a good approximation as the half lives of $^{221}\text{Fr}$ and $^{217}\text{At}$ are short enough so secular equilibrium with $^{225}\text{Ac}$ is ensured]
$u(R_{226\text{Ra}})$	= Standard uncertainty of net count rate for $^{226}\text{Ra}$ , in cpm
$R_{226\text{Ra}}$	= Net count rate for $^{226}\text{Ra}$ , in (cpm)

**NOTE: The uncertainty of half-lives and abundance values are a negligible contributor to the combined uncertainty and are considered during the evaluation of combined uncertainty.**

- A4.13. Calculate the mean and standard deviation of the mean (standard error) for the replicate determinations, to determine the acceptability of the tracer solution for use. The calculated standard deviation of the mean should be equal to or less than 5% of the calculated mean value.
- A4.14. Store the centrifuge tube containing the  $\text{Y}(\text{OH})_3/\text{Th}(\text{OH})_4$  precipitate. After sufficient time has elapsed a fresh  $^{225}\text{Ra}$  tracer solution may be generated by dissolving the precipitate with 40 mL of 0.5M  $\text{HNO}_3$  and repeating Steps A4.4 through A4.10 of this Appendix.