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Determination of radium-226 in drinking water by alpha spectroscopy

Jae-Gu Choi

University of Nevada, Las Vegas

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Determination of Ra-226 in drinking water by alpha spectroscopy

Choi, Jae-Gu, M.S.
University of Nevada, Las Vegas, 1991
DETERMINATION OF Ra-226 IN DRINKING WATER

BY ALPHA SPECTROSCOPY

by

Jae-Gu Choi

A thesis submitted in partial fulfillment of the requirements for the degree of

Master of Science

in

Environmental Analytical Chemistry

Chemistry Department
University of Nevada, Las Vegas
August, 1991
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ABSTRACT

A radiochemical procedure for determination of radium-226 (Ra-226) in drinking water was developed. The method involves concentrating radium by coprecipitation with barium sulfate; separating radium from barium and other alkaline earth metals by ion exchange; and electrodepositing radium onto stainless steel disc for radioactivity counting. The procedure requires the using a chemical yield tracer, Ra-223. Two methods for the preparation of Ra-223 from Ac-227 were studied. One method, which uses solvent extraction of radium with TTA/benzene, showed better results and took much less time in comparison to the other method which uses anion exchange, followed by co-precipitation of radium as Ba(Ra)SO₄ and dissolution with DTPA [7]. The results from the new analytical procedure for measuring Ra-226 in drinking water were compared with the traditional radon de-emanation method. The new procedure unfortunately showed no significant benefit over the de-emanation method.

Electrodeposition is a useful technique for preparing trace amounts of alpha-emitting radionuclides for radioactivity counting. The optimal conditions for electroplating several alpha-emitters, including radium-226 (Ra-226), uranium-238 (U-238), polonium-208 (Po-208), thorium-228 (Th-228), plutonium-239 (Pu-239), americium-241 (Am-241), and curium-
onto stainless steel discs from aqueous ammonium acetate were studied. The effects of varying the ammonium acetate concentration, pH, and the electroplating time were examined. The greatest chemical yield for Ra-226 occurred at a 0.175 M ammonium acetate solution with a pH of about 5, and a four-hour electroplating time; for Po-208, the greatest chemical yield occurred for a 0.175M ammonium acetate, a pH of 1, and a two-hour electroplating time. The optimal conditions for U-238 and Th-228 were a 0.35M ammonium acetate, a pH of 1 through 4, and a two-hour electroplating time. The optimal conditions for Pu-239, Am-241, and Cm-243,244 were at a 0.175M, a pH of 1 through 4, and about a two-hour electroplating time.
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to Jung-Hae Choi
my loving and supportive wife
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I. Introduction

Determination of Radium

The maximum permissible concentrations of radium-226 (Ra-226), radium-228 (Ra-228), and thorium isotopes in drinking water are controlled by Federal regulations. The regulations require that if the Ra-226 activity exceeds 3 picocuries per liter (pCi/L), the Ra-226 activity must be measured. If the combined activities of Ra-226 and Ra-228 exceed 5 pCi/L, the water supply exceeds the U.S. Environmental Protection Agency (USEPA) limit for radium in drinking water [1]. The determination of the naturally occurring radium isotopes (Ra-226 and Ra-228) in public and private water supplies has been the subject of many papers.

The most common method for determining radium-226 is the radon emanation method [13]. The technique involves collection of radon-222, the daughter of radium-226. The emanation method is relatively slow, requiring a waiting period of 20 days to obtain full equilibrium between the daughter Rn-222 and the parent, Ra-226.

Koide and Bruland[6] have developed a method for the spectroscopic determination of Ra-226 and other members of the U-238 series in sea water following precipitation of the
elements of interest with aluminium phosphate followed by precipitation with lead nitrate, ion exchange and electrodeposition onto a platinum disc for alpha countings. This method, however, is rather tedious, expensive, and uses corrosive fuming nitric acid.

Harada and Tsunogat [7] found that radium could be electroplated onto a silver disc from a hydrochloric acid solution during the radiochemical determination of thorium. A near quantitative electrodeposition of radium onto stainless steel from an ammonium acetate and nitric acid electrolyte between pH 8 and 9 was developed by Roman [8].

A technique described by Michel, Moor, and King [4] involves the preconcentration of radium in the field from 100 to 1000 L onto Mn-impregnated acrylic fiber cartridges. Radium is subsequently leached from the fiber by reducing the MnO₂ with hot HCl or desorbing the radium into cold diluted HNO₃. Radium is subsequently collected by coprecipitating with BaSO₄ and Ra-226 quantified by radon emanation.

Methods of determining Ra-228 published by USEPA involve either the extraction and counting of Ac-228, the short-lived daughter of Ra-228, or a sequential counting procedure which monitors the ingrowth of Ac-228 into a purified radium
sample [2]. A similar technique is described by Percival and Martin[3] which allows for the sequential determination of Ra-226, Ra-228, Ac-227, and thorium isotopes. However, this technique includes extensive wet-chemical separations and purifications which require skilled laboratory technique to produce consistent yield.

A alpha-gamma/beta-gamma coincidence counting technique has been reported to be a practical method for determining both Ra-226 and Ra-228 in water. However, the widespread application of this method may be slow because it requires a specialized radioactive detector.

Electrodeposition

There are very few reports on the electrodeposition of radium. A study of the electrolysis of radioferrous barium salts in acetone for the preparation of thin sources was published in 1937 [9]. Sorret and Tauveron [10] assumed that radium could not be satisfactorily electroplated on metal electrodes from aqueous solutions and instead adapted the Getoff and Bildstein plutonium technique and electrodeposited radium on platinum from isopropanol and HCl mixtures at 600 V and 12 mA [11]. Koide and Bruland used platinum discs and isopropanol as the
electrolyte effecting the electrodeposition at 40 V and 100-150 mA [6].

Roman [12] reported the electrodeposition of radium on stainless steel discs in high yields from aqueous ammonium acetate solution at pH 9. This method has advantages over the isoprophyl alcohol method in that the electrodeposition is effected from aqueous solutions at low voltage and moderately high current of 10-12 V and 400 mA, and a thin, strongly adhering film containing the radium is obtained which requires no fixing and which yields high resolution alpha-spectra.

Scope of the Thesis Research

There were two objectives in this research. The first objective was to improve the method for the determination of the activity of Ra-226 in drinking water, using Ra-223 as a chemical yield tracer. This research included 1) the separation of Ra-223 from Ac-227 by solvent extraction with TTA/benzene and cation exchange, 2) coprecipitating Ra-223 as Ba(Ra) SO₄, followed by anion exchange and cation exchange, 3) determination of Ra-223 by electrodeposition onto stainless steel disc with alpha spectroscopy, and 4) determination of Ra-226 in drinking water samples, including the separation of Ra-
226 from barium in the coprecipitate, Ba(Ra)SO₄, by cation exchange, followed by electrodeposition for determining activity of Ra-226. Results of this method for the determination of radium in drinking water were compared to results using the classical de-emanation method.

The second objective was to find the optimal conditions - concentration of ammonium acetate, pH, and electroplating time- for the electrodeposition of Ra-226, U-238, Po-208, Th-228, Pu-239, Am-241, and Cm-244 from aqueous ammonium acetate solution onto stainless steel discs.
II. Experimental Section

General

Apparatus

- Electroplating Cell (Figure 1); 20 ml polyethylene liquid scintillation vial with the bottom cut off, 3/4-inch stainless steel disc pressed firmly to the mouth of the vial with a modified cap as the cathode, and a platinum loop anode (100 g, 9 cm in length) inserted through the cut-off bottom into plating solution 12 mm from the cathode.
- Scintillation Vials: 20 ml, glass, Kimble.
- Automatic pipets: 100μ, 200μ, and 1000μ (Eppendorf).
- Ion Exchange Column size: 0.6 cm i.d., 10 cm long.

Instruments

- Alpha Counter: EG&G ORTEC, model 576A dual alpha spectrometer with model TR-SNA-450-100 semiconductor radiation detector, active area 450 mm² interfaced to Nuclear Data 6700 VAX/VMS.
- Gamma Counter: a 3.5 cm diameter by 7.1 cm deep well in a 10x10 cm NaI(Tl) gamma-ray detector (Cambera) interfaced to Nuclear Data 9900 VAX/VMS.
- Alpha Scintillation Cell: Lucas cell, model ASC-125, Volume 125 ml, Randam Electronic Inc. (Figure 2)
- Ludlum: model 2000 portable scalar rate meter (Figure 3)

Reagents

Reagent grade chemicals and deionized water (14 MΩ) were used in the preparation of all analytical solutions.

- Anion-Exchange Resin: Bio-Rad AG1-X8, 100-200 mesh.
- Cation-Exchange Resin: Bio-Rad AG 50W-X8, 100-200 mesh, hydrogen form.
- Isopropyl alcohol, 90 percent, Fisher.
- Hydrochloric acid, 1.5M, Fisher: Dilute 124.5 ml of the 37 percent reagent grade HCl to 1 liter with distilled water.
- Hydrochloric acid, 8M, Fisher: Dilute 664 ml of the 37 percent grade HCl to 1 liter with distilled water.
- Sulfuric acid, 18M, 96 percent H₂SO₄ reagent, Fisher.
- Ammonium Acetate Solution, Fisher: from pH 1 to pH 9 at each 0.175M, 0.35M, & 0.70M.
- Nitric acid, reagent A.C.S., Fisher
- Sodium hydroxide, 98.3 percent, Fisher
- 0.2M TTA/B (Thenoyltrifluoro Acetone/Benzene), 99 percent, Aldrich Chemical Company, Inc.
- DTPA (diethylenetriaminepentaacetate solution, 0.17M, pH 10.6): Dissolve 67g of diethylenetriaminepentaacetic acid (DTPA, 97%, Aldrich Chemical Company Inc) and 32g of sodium hydroxide pellets in 400 ml of water. Filter through double glass-fiber filters (0.45μ) with suction and dilute to 1 liter with water. Adjust the pH of 10 with sodium hydroxide or acid. Store the solution in a polyethylene bottles.

- Barium Carrier: 20 mg Ba++ /ml. Dissolve 35.6 grams of BaCl₂ 2H₂O (Baker) in 400 ml distilled water and dilute to 500 ml.

Radioactivities

- Ra-226 was purchased from Amersham/Searles Corporation.
- Ac-227 was purchased from Amersham/Searles Corporation.
- Ra-223 was separated from Ac-227.

All radioactive materials below were obtained from U.S. EPA-Environmental Monitoring Systems Laboratory-Las Vegas Nuclear Radiation Assessment Division with reference numbers that are traceable to NIST.

- Ra-226: #2557-2, 2430 dpm/ml, 0.5N-HCl, Dec.5,1989
- Po-208: 1.97 nCl/g, 2N-HCl, July.13,1988
- Am-241: #2344-3, 195.89 nCl/g, July 11, 1990
- Cm-244: SRM #4339, 69.50 Bq/g, 0.5N-HCl, June 13, 1984
- U-238: #1843-1, 2.22 nCl/g
- Th-228: RS#796, 654.23 dpm/g, May 23, 1990
- Ba-133 (#2529-1, 30579.24 dpm/ml) was obtained from the EPA- EMSL-LV.
Figure 1. Electroplating Cell
Procedure
1. Vacuum a cell
2. Purge with helium gas
3. Record the second bubbling time
4. Stay for 4 hr for equilibrium
5. Count

Cell Cleaning
1. 2 or 3 hr air cleaning
2. 30 min helium cleaning
3. Count for background

Figure 2. Instruments for radon-emanation method
Figure 3. Alpha scintillation counter for counting radon-222
Preparation of Ra-223 from Ac-227.

Figure 4. Decay chain shows parent/daughter relationship.

Tracer Ra-223 was prepared by milking a Ac-227 (half life: 21.6 years) solution that has equilibrated with Ra-223 (half-life 11.43 day) (Figure 4). Two methods were employed for Ra-223 preparation. The first one involved coprecipitating radium as Ba(Ra)SO₄. The coprecipitate was dissolved in DTPA (diethylenetriaminepentaacetic acid) and radium was subsequently purified by anion exchange and separated from barium through cation exchange.
Preparation of Ra-223 from Ac-227 by co-ppt. & ion exchange.

1) One ml of Barium carrier BaCl₂ was added to small portion (about one 1 ml) of Ac-227 with an activity of 68,240 dpm/ml which has been fully equilibrated with its daughter Ra-223. Two ml of 18M H₂SO₄ were also added into the solution. The solution was cooled to room temperature to allow the white precipitate, Ba(Ra)SO₄, to settle. The mixture was then centrifuged for 15 min.
The supernatant containing Ac-227 was decanted and saved for future milking of Ra-223. One ml of deionized water was added to wash the precipitate, shaken vigorously and centrifuged. The supernatant was decanted and 10 drops of 0.17M DTPA were added to the precipitate. The mixture was thoroughly mixed and placed into a water bath (70°C) until the precipitate dissolved completely.

Eight molar HNO$_3$ was added to bring the total volume to 10 ml. The solution was passed through an anion-exchange column that was packed with anion exchange resin (Bio-Rad AG1-X8, 100-200 mesh) preconditioned with 8M HNO$_3$. About 10 ml of the eluant was passed through a second anion-exchange column with the same resin conditions. About 10 ml of the eluant from the second column was evaporated to dryness and converted to the chloride with 12 M HCl and evaporated to dryness. The residue was dissolved with 10 ml of 1.6N-HCl, spiked with 1 ml of Ba-133. The solution was transferred to a glass scintillation vial and gamma counted to confirm the peaks of Ra-223 and Ba-133.

A cation-exchange column (Bio-Rad AG 50W-X8, 100-200 mesh) was prepared and preconditioned with 1.6N-HCl. The solution containing Ra-223 and Ba-133 was loaded onto the cation-exchange column and subsequently 120 ml of 1.6N-HCl was passed through the column. Each 10 ml of eluant was
collected in 20 ml glass scintillation vials and gamma counted. Vials were collected until Ba-133 was eluted completely.

v) Once the Ba-133 had eluted, the eluant was changed from 1.6N-HCl to 8N-HCl to remove Ra-233. The eluant containing the Ra-223 was collected in 150 ml a beaker and evaporated to dryness on a hot plate. The residue was dissolved with deionized water and evaporated to dryness. The residue was dissolved with 0.5N-HCl and brought to a final volume to 25 ml.

vi) This 25 ml solution was saved for future Ra-223 activity determination by electrodeposition against a known amount of Ra-226.
Ac-227 (Th-227, Ra-223)

\[ \text{BaCl}_2, \text{H}_2\text{SO}_4 \]

centrifuge

Supernatant (Ac-227)

Ba(Ra)SO_4

add water

centrifuge

Supernatant (Ac-227)

Ba(Ra)SO_4

dissolve with DTPA

8N-\text{HNO}_3

Anion Exchange Column

on column

eluent(Ra,Ba)

1.6N-\text{HCl}, \text{Ba-133}

Cation Exchange Column

1.6N-\text{HCl}

Ba, Ba-133

Ra-223
The second method of milking Ra-223 from Ac-227 employed solvent extraction with TTA/benzene (thenoyltrifluoroacetone/benzene) [6]. Ra-223 was extracted from the Ac-227 after Th-227 was stripped from the Ac-227 solution. Ra-223 was purified by cation-exchange.

TTA forms strong complex with many metal ions, particularly those of high valence.

Preparation of Ra-223 from Ac-227 by TTA extraction.

i) A small portion (about 1 ml) of Ac-227, activity of 68,240 dpm/ml which has been fully equilibrated with its daughter Ra-223 was added to 9 ml of deionized water and the pH was adjusted to a value of 2 with 0.1 M-HCl. The solution was extracted with 10 ml of 0.2 M TTA/benzene in a 125 ml separatory funnel. Th-227 is extracted into the organic phase at this pH.

ii) The pH value of the aqueous phase in the separatory funnel was adjusted to 5.5-6.0. Another 10 ml TTA/benzene was added to extract the Ac-227 into the organic phase. The Ac-227 was saved for future milking by back extracting the organic phase with 2N-HNO₃.
iii) The aqueous portion, which contains the Ra-223 was evaporated, to dryness. About 0.5 ml of conc. HCl was added and evaporated to dryness. The residue was dissolved with 2 ml of 1.5N-HCl and loaded onto a cation exchange resin column containing Bio-Rad AG50W-X8, 100-200 mesh preconditioned with 1.5N-HCl. An additional 120 ml of 1.5N-HCl was passed through the column. The Ra-223 adsorbed on the cation-resin and was subsequently eluted with 50 ml of 8N-HCl. The Ra-223 portion was collected in a 100 ml beaker and taken to near dryness. The residue in the beaker was dissolved with 1 ml of 0.5 N-HCl and diluted with 0.5 N-HCl to a total volume of 25 ml. This 25 ml solution containing the Ra-223 was saved to be used as the spiking solution.
Ac-227 (Th-227, Ra-223)

- add water (10 ml)
- adjust pH 2
- add 0.2M TTA/B (10 ml)

Solvent extraction

TTA/B (Th-227)

aqeuos (Ac-227, Ra-223)

- add TTA/B (10 ml)
- adjust pH 5.5-6.0

Solvent extraction

TTA/B (Ac-227)

- aqueous (Ra-223)

Stripping with 2N-HNO₃

(Ac-227)

Purification

Cation Exchange Column

1.6N-HCl

8N-HCl

Eluant (Ra-223)
Determination of Ra-223 activity.

Alpha-spectroscopy was used to determine the activities of Ra-223 obtained from Ac-227.

**Direct deposition onto a platinum disc.**

One hundred µl of the Ra-223 spiking solution and fifty µl of known Ra-226 activities were placed directly onto a platinum disc (i.d. 2 cm) and allowed to evaporate to dryness and then flamed. The disc was counted by alpha-spectroscopy for 3 min. The activity of Ra-223 was calculated by comparing it to the Ra-226 activity.

\[
\text{Conc. of Ra-223} = \frac{\text{Conc. of Ra-226 spike} \times \frac{T.C. \text{ of Ra-223}}{T.C. \text{ of Ra-226}} \times F}{- T.C.; \text{Total Counts}}
\]

- T.C.: Total Counts
- F: Correction Factor (Decay Factor)

**Electrodeposition.**

Five hundred µl of the Ra-223 spiking solution and two hundred µl of known Ra-226 were introduced into an electroplating cell that had been assembled for
electrodeposition. Then 5 ml of 90 percent isopropyl alcohol and 0.5 ml of 0.05 M-HCl were added to the electroplating cell (Figure 1). A 1/4 inch carbon rod or a platinum rod was used as the anode. The voltage was adjusted to 40 volts yielding a current of from 20-40 mA and electroplated for one hour. After plating, the electrodeposition all was washed with isoprophyl alcohol and disassembled. The stainless steel disc containing the Ra-223 and Ra-226 was counted in an alpha counter for 30 min. The Ra-223 activity was calculated by the following equation.

\[
\text{Conc. of Ra-223} = \frac{\text{Conc. of Ra-226 spike} \times \frac{\text{T.C. of Ra-223}}{\text{T.C. of Ra-226}} \times F}{\text{T.C.}}
\]

- T.C.; Total Counts
- F: Correction Factor (Decay Factor)

**Determination of Ra-226 in drinking water sample**

Barium-133 and Ra-223 were spiked into water samples along with stable barium carrier. Sulfuric acid was then added to precipitate the barium sulfate. The precipitate was allowed to settle, collected, washed, and dissolved with DTPA. Cation exchange (Bio-Rad AG50W-X8, 100-200 mesh) was used to separate the barium carrier from the radium because barium has a lower affinity for the resin in 1.6 M HCl, as described in the
Ra-223 section. The complete separation was determined when barium-133 was no longer found in the eluant. Radium was then eluated from the column with 8 N-HCl. Its progress was monitored by the gamma activity of Ra-223.

Procedure for determination of Ra-226 in drinking water sample

1) Two hundred ml of drinking water sample (tap water) was spiked with one ml of Ra-223 (about 50 dpm/ml), 2 ml of BaCl₂ (20 mg Ba⁺⁺ per ml) and one ml of Ba-133 (about 10,000 dpm/ml). The solution was evaporated to about 5 ml. Two ml of conc. H₂SO₄ were added and mixed vigorously. At this time, a white precipitate of Ba(Ra)SO₄ appeared. The solution was cooled to room temperature.

2) The precipitate was collected by centrifugation. The supernatant was decanted and one ml of 0.17M DTPA added to the precipitate. The precipitate dissolved completely at 70°C. The dissolved precipitate was loaded onto a AG50X8 (100-200 mesh) cation exchange column. Five ml of NaOH (pH value of 10.5) were subsequently passed through the column.

3) One hundred twenty ml of 1.6N-HCl were passed through to elute the barium. The radium fraction, containing Ra-223 and Ra-226 was removed from the resin with 60 ml 8N-HCl. The
eluant was evaporated to dryness on the hot plate. One half ml of 0.5N-HCl was added to dissolve the residue. Five ml of isopropyl alcohol was added. The mixture was then quantitatively transferred to an electroplating cell. After electroplating for one hour, the stainless steel planchet was counted for 1000 min in an alpha counter. The Ra-226 activity was calculated with the Ra-223, using the yield by the following formula:

\[
\frac{\text{net cpm}}{E \times 2.22 \times R \times L_{\text{sample}}} = \text{Ra-226 (pCi/L)}
\]

- net cpm = gross cpm - background cpm.
- L = volume of the sample in L
- R = radiometric recovery of Ra-223
- E = alpha detector efficiency
Drinking Water Sample

1. Evaporate
2. Add BaCl₂, Ba-133, Ra-223, evaporate H₂SO₄
3. Centrifuge
4. Add water
5. Centrifuge
6. Dissolve with DTPA
7. 1.6N-HCl
8. Cation Exchange Column
9. 1.6N-HCl
10. Ba on column
11. 8N-HCl
12. Ra-223, 226
13. Go to electrodeposition
**Rn-Emanation Method**

Ra-226 was also determined by the de-emanation method [13], using radon bubblers and radon scintillation cells described by Lucas [14]. The samples were USEPA-EMSL-LV Cross-Check Sample, type: radium in water (11-Mar-88, 17-June-88, 16-Sep-88, 10-Mar-89, 14-Jul-89, 10-Nov-89, 09-Mar-90, and 13-Jul-90).

![Rn-222/Ra-226](image)

**Figure 6. Equilibrium of Ra-226 and its daughter Rn-222**
Procedure for de-emanation method

1) Two ml of Ba/Pb carrier (20 mg/ml, 35.6g BaCl₂ 2H₂O/L) and 2 ml of Pb(NO₃)₂ (20 mg/ml) were added to 200 ml U.S. EPA EMSL-LV cross check sample (Table 3). The solution was heated to boiling. Twenty ml of 9 M H₂SO₄ was added very carefully. The solution was heated for 10 min and cooled to room temperature. The solution was transferred to a 500 ml centrifuge bottle and centrifuged for 15 min. The supernatant was discarded. The precipitate was transferred into a 50 ml centrifuge tube with 0.05N H₂SO₄. The mixture was centrifuged for 10 min. and the supernatant discarded.

2) The precipitate was washed with 5 ml of 0.05N H₂SO₄ and centrifuged for 10 min. The supernatant was discarded. The precipitate was dissolved with approximately 10 ml 0.17M DTPA at 70°C. The solution was transferred to a radon bubbler (Figure 2).

3) The upper and lower stopcocks were opened to purge all gases from the solution by slowly passing helium gas through the bubbler for about 20 min.

4) The stopcocks were closed, and the time recorded. The solution was then stored for 14 to 20 days for the ingrowth of Rn-222 (Figure 6).
v)  At the end of the storage period, the upper half of an absorption tube was filled with magnesium perchlorate and the lower half filled with ascarite.

vi) The tube was attached to the radon bubbler and then attached to the evacuated scintillation cell. The assembly was checked for leaks. The outlet stopcock on the bubbler was opened, and when no further significant bubbling took place, the stopcock was closed.

vii) The helium gas pressure was adjusted so that the gas flowed at slightly above atmospheric pressure. The hose to the bubbler inlet was connected the inlet stopcock gradually opened using the bubbling as a guide. When the stopcock was opened gradually without a significant amount of bubbling, the bubbler was essentially at atmospheric pressure again.

viii) The outlet stopcock was opened very slightly to allow bubbling to proceed at a rate, determined by experience, such that 15 to 20 min. were required to complete de-emanation (purging of radon).

ix) Toward the end of the de-emanation, the helium gas pressure was gradually increased. When the system was at atmospheric pressure, the helium gas was shut off, the tubing was disconnected from the bubbler inlet and the inlet and outlet
stopcocks closed. This marked the beginning of Ra-222 decay and ingrowth of Ra-222 daughters.

x) The scintillation cell was stored for at least 4 hours to ensure equilibrium between radon and radon daughters.

xi) The activity in the alpha scintillation cell was counted in a Ludlum 2000 portable rate meter.

Equation for Ra-226 calculation

\[
Ra-226 \text{ (pCi/L)} = \left( \frac{C_l - C_b}{2.22 \times E \times V} \right) \\
\times \left[ \frac{1}{1-e^{-\lambda t_1}} \right] \\
\times \left[ \frac{1}{1-e^{-\lambda t_2}} \right] \\
\times \left[ \frac{\lambda t_3}{1-e^{-\lambda t_3}} \right]
\]

C_l: Gross count
C_b: Background
E: Calibration factor for Lucas cell
V: Sample volume in Liter
\( \lambda \): Decay constant for Rn-222
\( t_1 \): Time for in-growth of Rn-222
\( t_2 \): Time between de-emanation and counting
\( t_3 \): Counting time
2.22 = dpm/pCi
Electrodeposition

To determine the optimal electroplating conditions for a variety of radionuclides, 0.175M, 0.35M, and 0.70M concentrations of ammonium acetate, pH 1 through pH 9 for each ammonium acetate concentration, and one hour to four hours electroplating time were studied.

<table>
<thead>
<tr>
<th>Ammonium Acetate Solution</th>
<th>Ra-226</th>
<th>Po-208</th>
<th>U-238</th>
<th>Th-228</th>
<th>Pu-239</th>
<th>Am-241</th>
<th>Cm-244</th>
</tr>
</thead>
<tbody>
<tr>
<td>Conc.</td>
<td>0.17M</td>
<td>0.17M</td>
<td>0.17M</td>
<td>0.17M</td>
<td>0.17M</td>
<td>0.17M</td>
<td>0.17M</td>
</tr>
<tr>
<td></td>
<td>0.35M</td>
<td>0.35M</td>
<td>0.35M</td>
<td>0.35M</td>
<td>0.35M</td>
<td>0.35M</td>
<td>0.35M</td>
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<tr>
<td></td>
<td>0.70M</td>
<td>0.70M</td>
<td>0.70M</td>
<td>0.70M</td>
<td>0.70M</td>
<td>0.70M</td>
<td>0.70M</td>
</tr>
<tr>
<td>pH</td>
<td>1 to 9</td>
<td>1 to 9</td>
<td>1 to 9</td>
<td>1 to 9</td>
<td>1 to 9</td>
<td>1 to 9</td>
<td>1 to 9</td>
</tr>
<tr>
<td>Electroplating time</td>
<td>1 to 4 hr</td>
<td>1 to 4 hr</td>
<td>1 to 4 hr</td>
<td>1 to 3 hr</td>
<td>1 to 4 hr</td>
<td>1 to 4 hr</td>
<td>1 to 4 hr</td>
</tr>
</tbody>
</table>

Table 1. Combinations of electroplating conditions studied

The electroplating cell and the platinum electrode were soaked in 1N-HNO₃ overnight and then washed with deionized water. The electroplating cell was assembled with a stainless steel disc as cathode (see Figure 1).
i) Ten ml of ammonium acetate solution were added as the electrolyte to the cell and one of the target radionuclides was introduced. The electrolysis was carried out at a current of 400 mA which required a voltage of about 12V. Evaporation of the electrolyte caused by the heat produced during plating was compensated by adding a few drops of ammonium acetate solution from time to time. After electrolysis was finished, the stainless steel disc was washed with acetone.

ii) The stainless steel disc with the radionuclide plated on its polished side was mounted in an alpha-counter and counted for about 30 minutes. This chemical yield for each radionuclide for each combination of the electroplating conditions was determined.
III. Results and Discussion

Preparation of Ra-223 from Ac-227

The proposed chemical yield tracer, Ra-223 (half life, 11.7 d) could be separated repeatedly (milked) from Ac-227 (half life, 21.6y) in sufficient activity to serve as a useful tracer for about two months. The TTA/benzene extraction technique to prepare Ra-233 from Ac-227 was found to be superior to the method that uses barium sulfate coprecipitation followed by ion exchange. The ion-exchange method required three times more time than the extraction method, three days versus one day, even though the activities of Ra-223 obtained by two methods are insignificantly different.

Separation of Ra-223,226 from Ba-133

Coprecipitation of radium from water by the barium sulfate procedure produces a precipitate from which radium must be separated from the macroamount of added barium. Ra-223 and Ba-133 which were spiked into water sample are gamma emitters, and were used to monitor the progress of this separation. A clean separation of radium from barium (Figure 7) was accomplished by cation-exchange by monitoring the gamma
activity of each 10 ml aliquot of 1.6N-HCl which was used as the separating medium.

Barium was completely eluted from the cation exchange resin column after passing between 130 ml 1.6N-HCl and 240 ml 1.6N HCl, while radium adhered to the resin (Figure 7). Ra-223,226 were subsequently eluated with 60 ml of 8N HCl.

Figure 7. The separation of Ra-223 from Ba-133.
Preconcentration of radium from drinking water

The average recoveries of Ra-223 by coprecipitation with barium sulfate, the first step in the method, was found to be 89% ± 5% indicating good scavenging of radium from drinking water by barium sulfate.

Comparison of electrodeposition (new procedure) with de-emanation method (old procedure).

<table>
<thead>
<tr>
<th>SAMPLE#</th>
<th>EMANATION</th>
<th>ELECTRODEPOSITION</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>88%</td>
<td>51%</td>
</tr>
<tr>
<td>2</td>
<td>99%</td>
<td>46%</td>
</tr>
<tr>
<td>3</td>
<td>87%</td>
<td>60%</td>
</tr>
<tr>
<td>4</td>
<td>87%</td>
<td>52%</td>
</tr>
<tr>
<td>5</td>
<td>89%</td>
<td>31%</td>
</tr>
<tr>
<td>6</td>
<td>85%</td>
<td>78%</td>
</tr>
<tr>
<td>7</td>
<td>95%</td>
<td>46%</td>
</tr>
</tbody>
</table>

†The media for electrodeposition was Ammonium Acetate solution.
Table 2. Comparison of electrodeposition with de-emanation method

Results from the new method utilizing Ra-223 as a chemical yield monitor, were compared to the classical de-emanation method. Even though de-emanation method takes more than 20 days to get full equilibrium between parent Ra-226 and daughter Rn-222, the Rn-emanation method with 85% to 99% recoveries is better than new procedure with 31% to 78% recoveries.
Comparison of aqueous ammonium acetate with isopropyl alcohol as a medium for electrodeposition

After an extensive investigation of aqueous ammonium acetate as a medium for the electrodeposition of radium onto stainless steel, the average recovery of 77% ± 5% was found to occur at an ammonium acetate concentration of 0.35 M, a pH of 8, and a plating time of four hours. However, radium was found to plate onto stainless steel in recoveries of 98% ± 3% from 90% isopropyl alcohol and 10% 0.05 M HCl in a plating time of one hour (Table 2). Therefore, isoprophyl alcohol instead of ammonium acetate solution was used as a media for electrodeposition of Ra-223,226 for determining their activities.
Comparison of electrodeposition for ammonium acetate and isopropyl alcohol

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>I.D.</th>
<th>Chem. Yield of Ra-223</th>
<th>Corrected Activity of Ra-226</th>
<th>True Activity of Ra-226</th>
<th>Sol'n Type</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>17-Jun-88</td>
<td>0.76%</td>
<td>8.77pCi/L</td>
<td>10.0pCi/L</td>
<td>AA</td>
</tr>
<tr>
<td>2</td>
<td>16-Sep-88</td>
<td>1.20%</td>
<td>1.61pCi/L</td>
<td>8.4pCi/L</td>
<td>AA</td>
</tr>
<tr>
<td>3</td>
<td>13-Jul-90</td>
<td>0.18%</td>
<td>9.06pCi/L</td>
<td>12.1pCi/L</td>
<td>AA</td>
</tr>
<tr>
<td>4</td>
<td>9-Nov-90</td>
<td>0.19%</td>
<td>5.58pCi/L</td>
<td></td>
<td>AA</td>
</tr>
<tr>
<td>5</td>
<td>17-Jun-88</td>
<td>7.28%</td>
<td>7.92pCi/L</td>
<td>10.0pCi/L</td>
<td>I.S</td>
</tr>
<tr>
<td>6</td>
<td>16-Sep-88</td>
<td>6.88%</td>
<td>5.72pCi/L</td>
<td>8.4pCi/L</td>
<td>I.S</td>
</tr>
<tr>
<td>7</td>
<td>13-Jul-90</td>
<td>6.76%</td>
<td>10.3pCi/L</td>
<td>12.1pCi/L</td>
<td>I.S</td>
</tr>
<tr>
<td>8</td>
<td>9-Nov-90</td>
<td>9.50%</td>
<td>7.13pCi/L</td>
<td></td>
<td>I.S</td>
</tr>
</tbody>
</table>

Table 3. The results of electrodeposition with isopropyl alcohol and ammonium acetate solution (0.35M, pH 8) as media. Sample Type: radium in water; Isotope: Ra-226, Ra-228.
Electrodeposition

In the second part of the thesis research, the electrodepositing conditions for seven radionuclides were examined using aqueous ammonium acetate as a medium to determine the best yield conditions. All of these radionuclides are alpha emitters and their half lives and alpha energy levels are shown in Table 4. Uranium, plutonium, americium, and curium are waste products in nuclear power.

<table>
<thead>
<tr>
<th>Radionuclides</th>
<th>Half-life</th>
<th>Energy Level</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ra-226</td>
<td>1602yr</td>
<td>4782kev (95%)</td>
</tr>
<tr>
<td>Po-208</td>
<td>2.879yr</td>
<td>5115kev (100%)</td>
</tr>
<tr>
<td>U-238</td>
<td>$4.51 \times 10^9$ yr</td>
<td>4196kev (77%)</td>
</tr>
<tr>
<td>Th-228</td>
<td>1.910yr</td>
<td>5427kev (71%)</td>
</tr>
<tr>
<td>Pu-239</td>
<td>24,400yr</td>
<td>5156kev (73%)</td>
</tr>
<tr>
<td>Am-241</td>
<td>458yr</td>
<td>5486kev (86%)</td>
</tr>
<tr>
<td>Cm-244</td>
<td>17.6yr</td>
<td>5806kev (77%)</td>
</tr>
</tbody>
</table>

Table 4. The energy levels and half lives of radionuclides used for electrodeposition.

The greatest chemical yield (77% ± 5%) for Ra-226 occurred at a 0.175 M ammonium acetate solution with a pH of
about 5, and a four-hour electroplating time; for Po-208, the
greatest chemical yield 91% ± 5%) occurred for a 0.175M
ammonium acetate, a pH of 1, and a two-hour electroplating
time. The optimal conditions for U-238 and Th-228 were a
0.35M ammonium acetate, a pH of 1 through 4, and a two-hour
electroplating time. Their chemical yields were 100% ± 5%.
The optimal conditions for Pu-239, Am-241, and Cm-243,244
with yields of 85% ± 5%, 90% ± 5%, and 100% ± 5%
respectively, were at a pH of 1 through 4, and about a two-hour
electroplating time.

Optimal Conditions

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>conc. (A.A.)</th>
<th>pH</th>
<th>Plating Time(hr)</th>
<th>Chem. Yield</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ra-226</td>
<td>0.175M</td>
<td>5</td>
<td>4</td>
<td>77.5(%)</td>
</tr>
<tr>
<td>Po-208</td>
<td>0.175M</td>
<td>1</td>
<td>2</td>
<td>91.7(%)</td>
</tr>
<tr>
<td>U-238</td>
<td>0.35M</td>
<td>from 1 to 4</td>
<td>2</td>
<td>100(%)</td>
</tr>
<tr>
<td>Th-228</td>
<td>0.35M</td>
<td>1</td>
<td>2</td>
<td>100(%)</td>
</tr>
<tr>
<td>Pu-239</td>
<td>0.175M</td>
<td>from 1 to 4</td>
<td>2</td>
<td>85(%)</td>
</tr>
<tr>
<td>Am-241</td>
<td>0.175M</td>
<td>from 1 to 4</td>
<td>1</td>
<td>90(%)</td>
</tr>
<tr>
<td>Cm-244</td>
<td>0.175M</td>
<td>from 1 to 3</td>
<td>2</td>
<td>100(%)</td>
</tr>
</tbody>
</table>

† The media for electrodeposition was aqueous ammonium acetate (A.A).  
Table 5. The optimal conditions for seven natural radionuclides
by electrodeposition according to conc., pH, & electroplating
time.
Figures in next seven pages show the optimal conditions for seven radionuclides at different concentration of ammonium acetate solution, pH, and electroplating time. The other figures and data are attached to appendix.
0.175 M Ammonium Acetate

- - - - 4 hrs.
- - - - 3 hrs.
- - - - 2 hrs.
- - - - 1 hr.

pH vs. Ra-226
0.175 M Ammonium Acetate

- 4 hrs.
- 3 hrs.
- 2 hrs.
- 1 hr.

U-238 vs pH
0.175 M Ammonium Acetate

- - - 4 hrs.
- - 3 hrs.
- - 2 hrs.
- - 1 hr.

pH vs. Po-208

EL.0PH0/P10-1960 10
0.175 M Ammonium Acetate

- 4 hrs.
- 3 hrs.
- 2 hrs.
- 1 hr.

Cm-243, 244 vs. pH
References


0.70 M Ammonium Acetate

- - - - - 4 hrs.
- - - - - 3 hrs.
- - - - - 2 hrs.
- - - - - 1 hr.

Ra-226 vs pH

ELQR/PWP10-10-00 9
0.175 M Ammonium Acetate

- - - 3 hrs.
--- 2 hrs.
- - - 1 hr.

Th-228 vs pH graph
0.35 M Ammonium Acetate

- --- 4 hrs.
- - - 3 hrs.
- - - 2 hrs.
- --- 1 hr.
0.35 M Ammonium Acetate

- 4 hrs.
- 3 hrs.
- 2 hrs.
- 1 hr.

Pu-239 vs pH

ELGRPHS/P/10-10-90 20
0.70 M Ammonium Acetate

Pu-239 vs pH

- • • • 4 hrs.
- - - 3 hrs.
- .-- 2 hrs.
- -- 1 hr.

ELGPH/09/10-19-80 21
0.35 M Ammonium Acetate
- - - 4 hrs.
--- 3 hrs.
---- 2. hrs.
----- 1 hr.

Cm-243, 244

pH
0.70 M Ammonium Acetate

- 4 hrs.
- 3 hrs.
- 2 hrs.
- 1 hr.

Cm-243, 244

pH

1 2 3 4 5 6 7 8 9

0 10 20 30 40 50 60 70 80 90 100
## Ra-226

<table>
<thead>
<tr>
<th>Time/hr.</th>
<th>Conc./M</th>
<th>pH</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td>0.175</td>
<td>0.57 ± 0.3</td>
</tr>
<tr>
<td></td>
<td>0.35</td>
<td>0.22 ± 0.2</td>
</tr>
<tr>
<td></td>
<td>0.70</td>
<td>0.45 ± 0.2</td>
</tr>
<tr>
<td></td>
<td>0.175</td>
<td>3.41 ± 0.6</td>
</tr>
<tr>
<td></td>
<td>0.35</td>
<td>2.14 ± 0.5</td>
</tr>
<tr>
<td></td>
<td>0.70</td>
<td>1.12 ± 0.3</td>
</tr>
<tr>
<td></td>
<td>0.175</td>
<td>5.75 ± 0.9</td>
</tr>
<tr>
<td></td>
<td>0.35</td>
<td>11.4 ± 1.3</td>
</tr>
<tr>
<td></td>
<td>0.70</td>
<td>6.34 ± 0.9</td>
</tr>
<tr>
<td></td>
<td>0.175</td>
<td>12.8 ± 1.4</td>
</tr>
<tr>
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<td>0.35</td>
<td>14.0 ± 1.5</td>
</tr>
<tr>
<td></td>
<td>0.70</td>
<td>6.08 ± 0.9</td>
</tr>
<tr>
<td>Time/hr.</td>
<td>Conc./M</td>
<td>pH</td>
</tr>
<tr>
<td>---------</td>
<td>---------</td>
<td>-------------</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1</td>
</tr>
<tr>
<td>1</td>
<td>0.175</td>
<td>73.2±6.0</td>
</tr>
<tr>
<td></td>
<td>0.35</td>
<td>72.4±5.9</td>
</tr>
<tr>
<td></td>
<td>0.70</td>
<td>84.7±6.0</td>
</tr>
<tr>
<td>2</td>
<td>0.175</td>
<td>93.2±6.9</td>
</tr>
<tr>
<td></td>
<td>0.35</td>
<td>98.7±7.2</td>
</tr>
<tr>
<td></td>
<td>0.70</td>
<td>83.8±6.5</td>
</tr>
<tr>
<td>3</td>
<td>0.175</td>
<td>96.5±7.1</td>
</tr>
<tr>
<td></td>
<td>0.35</td>
<td>99.3±7.3</td>
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<td>0.70</td>
<td>88.2±6.8</td>
</tr>
<tr>
<td>4</td>
<td>0.175</td>
<td>98±7.9</td>
</tr>
<tr>
<td></td>
<td>0.35</td>
<td>110.3±7.9</td>
</tr>
<tr>
<td></td>
<td>0.70</td>
<td>83.6±6.5</td>
</tr>
</tbody>
</table>
## Th-228

<table>
<thead>
<tr>
<th>Time/hr.</th>
<th>Conc./M</th>
<th>pH</th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
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<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
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<td>2</td>
<td>3</td>
<td>4</td>
<td>5</td>
<td>6</td>
<td>7</td>
<td>8</td>
<td>9</td>
</tr>
<tr>
<td>0.175</td>
<td>58 ± 8.1</td>
<td>53 ± 8.5</td>
<td>45 ± 5.3</td>
<td>24 ± 3.7</td>
<td>21 ± 0.9</td>
<td>0.2 ± 0.3</td>
<td>0.18 ± 0.36</td>
<td>0.96 ± 0.6</td>
<td>1.77 ± 1.17</td>
</tr>
<tr>
<td>0.35</td>
<td>87.6 ± 8.3</td>
<td>63.7 ± 6.6</td>
<td>65.8 ± 7.5</td>
<td>26.5 ± 3.9</td>
<td>1.3 ± 0.7</td>
<td>0.86 ± 0.6</td>
<td>0.77 ± 0.6</td>
<td>0.19 ± 0.3</td>
<td>0.99 ± 0.61</td>
</tr>
<tr>
<td>0.70</td>
<td>59.5 ± 6.3</td>
<td>63.7 ± 6.7</td>
<td>51.3 ± 5.8</td>
<td>0.7 ± 0.5</td>
<td>0.3 ± 0.4</td>
<td>0.1 ± 0.4</td>
<td>0.18 ± 0.36</td>
<td>0.5 ± 0.4</td>
<td>0.18 ± 0.36</td>
</tr>
<tr>
<td>0.175</td>
<td>69.5 ± 8.3</td>
<td>70.9 ± 7.8</td>
<td>53.4 ± 5.9</td>
<td>23.5 ± 3.6</td>
<td>7.2 ± 1.8</td>
<td>2.3 ± 1.1</td>
<td>4.0 ± 1.3</td>
<td>0.9 ± 0.6</td>
<td>0.8 ± 0.6</td>
</tr>
<tr>
<td>0.35</td>
<td>106.8 ± 9.4</td>
<td>93.6 ± 8.6</td>
<td>72.9 ± 7.3</td>
<td>23.6 ± 3.6</td>
<td>8.4 ± 2.0</td>
<td>11.4 ± 2.4</td>
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**Am-241**

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