Ra-06-RC

RADIUM-226 IN URINE AND WATER

Contact Person(s) : Isabel M. Fisenne

APPLICATION

This procedure is applicable to samples of urine and water. Small amounts of thorium are carried by the $BaSO_4$ and the method fails in the presence of ^{230}Th. Natural thorium does not interfere. Other α-emitting isotopes of radium also interfere.

Radium is initially separated from untreated water or urine by coprecipitation with calcium phosphate. Calcium and most other elements are removed by coprecipitation of radium on barium sulfate. Organic material is removed by ignition, silica is removed by volatilization with HF, and the radium-barium sulfate is reprecipitated.

Radium-226 in the barium sulfate precipitate is stored to allow the buildup of ^{222}Rn, ^{218}Po, and ^{214}Po. The equilibrated α activities are then measured with a scintillation counter. The instrument response is converted to Bq by application of corrections for counter background and efficiency and for self-absorption. Chemical recovery is measured gravimetrically and the ^{226}Ra activity is equivalent to one-fourth of the total Bq measured.

SPECIAL APPARATUS

1. Rings and discs - see Specification 7.2.
2. Teflon filter holders and filter funnels and sample mounts - see Specification 7.12.
3. Mylar film - see Specification 7.10.

SPECIAL REAGENTS

1. Barium carrier solution: 5 mg Ba mL^{-1} - 9.51 g $Ba(NO_3)_2$ L^{-1} of 1:99 HCl.

SAMPLE PREPARATION

A. Water.

1. Transfer 4 L of the sample to a 5 L stainless steel pot.

2. Add 1 mL of barium carrier solution and 4 mL of HNO_3.

3. Evaporate to near dryness, add additional aliquots of water and HNO_3, and continue until 50 L of sample have been treated.

4. Evaporate to about 100 mL, add 3-4 g of NH_4Ac, and adjust the pH to 5 with 1:1 NH_4OH.

B. Urine.

1. Transfer a measured volume of urine to a 250-mL centrifuge bottle and add 1 mL of barium carrier solution.

2. Add 2 mL of H_3PO_4. Adjust the pH to 9 with 1:1 NH_4OH. Stir, centrifuge, and discard the supernate.

3. Dissolve the precipitate with 10 mL of HNO_3. Dilute to about 100 mL with water.

4. Add 1 mL of H_3PO_4. Adjust the pH to 9 with 1:1 NH_4OH. Stir, centrifuge, and discard the supernate.

5. Dissolve the precipitate in a few mL of acetic acid and dilute to 100 mL with water. Adjust the pH to 5 with 1:1 NH_4OH.

DETERMINATION

1. Add 3-4 g of $(NH_4)_2SO_4$ with stirring. Digest for 12 h. Cool and filter by gravity on a 9 cm Whatman No. 42 filter paper. Wash with 0.5% H_2SO_4. Discard the filtrate and washings.

2. Transfer the precipitate and filter to a platinum dish. Dry and ignite at 900°C.

3. Cool and add 2 mL of 1:1 H_2SO_4 and 5 mL of HF. Evaporate on a sand bath until SO_3 fumes appear.

4. Transfer the solution to a 40 mL, heavy walled centrifuge tube with 30 mL of H_2O and digest for 12 h. Centrifuge and discard the supernate.

5. Wash the precipitate with 10 mL of water. Centrifuge and discard the washings.

6. Slurry the precipitate with water and filter on a tared Whatman No. 42 filter paper.

7. Dry in an oven at about 150°C for at least 2 h.

8. Cool, weigh, and mount on a ring and disc with an α phosphor and Mylar.

9. Store for 30 days and count on an α-scintillation counter.

DATA PROCESSING AND ANALYSES

The ^{226}Ra disintegration rate is obtained from the counting rate of the equilibrated radium fraction through the following calculation:

$$Bq = R_s \, YTCE$$

where R_s is the net counting rate of the sample, Y is the recovery factor, T is the self-absorption correction, E is the counter efficiency factor, and C is a theoretical conversion from total α activity to ^{226}Ra α activity.

Chemical recovery, Y, is obtained by weighing the final Ba sulfate precipitate. The calculation of the recovery factor is:

$$Y = \frac{x}{(w - t)\, f}$$

where x is the weight of barium added as carrier, w is the total weight of the final precipitate and filter paper, t is the weight of the filter paper, and f is the gravimetric factor which equals 0.588 mg barium per mg of barium sulfate.

Self-absorption correction, T, is used to normalize the self-absorption of the ^{226}Ra, ^{222}Rn, ^{218}Po, and ^{214}Po α activities in samples and standards to a common thickness (mg cm^{-2}). T is obtained by counting representative α emitters over the energy range of 4.8-8.8 MeV in precipitates of varying sample thicknesses. The correction is taken as the ratio of counting rates at an arbitrary minimum thickness to other thicknesses over the range of probable sample recoveries. Figure 1 is a typical composite plot of the correction obtained for ^{230}Th, ^{212}Pb, and ^{226}Ra activities for varying thicknesses of their oxalate, chromate, and sulfate derivatives, respectively.

Theoretical activity conversion factor, C, equals 0.25 after 30 days of buildup, when the three α-emitting progeny are in secular equilibrium with ^{226}Ra. However, the ratio of ^{226}Ra to total α activity at any time after the separation of radium may be calculated from the Bateman equation for buildup of ^{222}Rn.

Efficiency factor, E, is determined by α counting a known quantity of ^{226}Ra under sample conditions. The correction is taken as the ratio of the theoretical activity to the observed counting rate. The calculation is expressed as:

$$E = \frac{A}{R_S' T' C'}$$

where A is the theoretical activity of the standard, and R_S' is the net counting rate of the standard, T' is the self-absorption correction for the standard, and C' is the theoretical ratio of ^{226}Ra to total α activity in the standard.

An accompanying computational data sheet illustrates a procedure for routine calculation of ^{226}Ra activity rates. Experimental data are tabulated as they are derived and the calculations are performed on a step-by-step basis.

LOWER LIMIT OF DETECTION (LLD)*

Counter efficiency	(%)	50
Counter background	(cps)	1.67×10^{-5}
Yield	(%)	85
Blank	(cps)	1.67×10^{3}
LLD (400 min)	(mBq)	3
LLD (1000 min)	(mBq)	2

*Reagent blank must be analyzed with each set of samples.

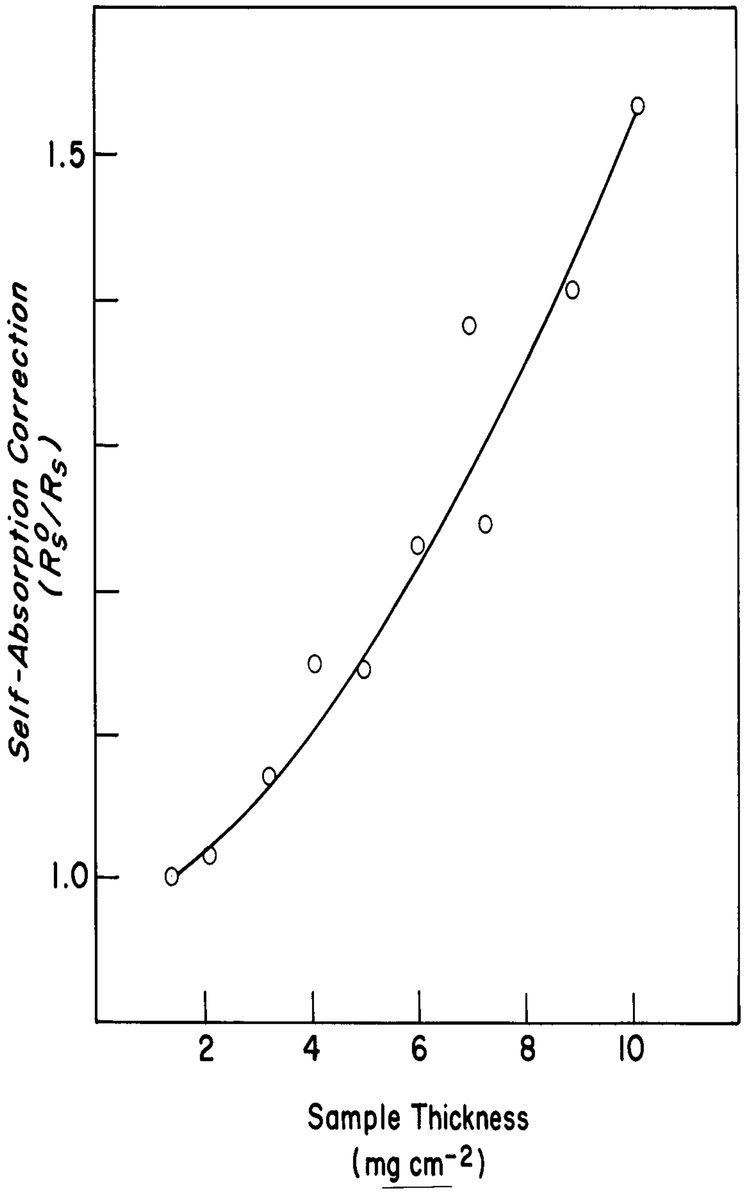

Figure 1. Self-absorption for α activities (4.8-8.9 MeV).

Date _________________ Sample Number

		Operation						
1	Counter number	-						
	STANDARD COUNTING							
2	Gross background count	-						
3	Count interval (s)	-						
4	Background (cps)	2/3						
5	Gross standard count	-						
6	Count time	-						
7	Standard (cps)	5/6						
8	Standard (net cps)	7-4						
	CORRECTIONS							
9	Gross weight (mg)	-						
10	Tare weight	-						
11	Net weight	9-10						
12	Weight of barium	11x0.59						
13	Carrier added	-						
14	Chemical yield factor (Y)	13/12						

Date ________________ Sample Number

		Operation						
11	Net weight	Copy						
15	Thickness correction (T)	Graph						
16	Corrected standard (cps)	8x14x15						
17	Added x 4* (Bq)	-						
18	Efficiency factor (E)	17/16						

*To include progenys

Date _________________ Sample Number

		Operation						
1	Counter number	-						
3	Count interval (s)	Copy						
4	Background (cps)	Copy						
18	Efficiency factor (E)	Copy						
	SAMPLE COUNTING							
19	Gross sample count	-						
20	Count time	-						
21	Sample (cps)	19/20						
22	Sample (net cps)	21-4						
23	Sample (Bq)	22x18						
	CORRECTIONS							
24	Gross weight (mg)	-						
25	Tare weight	-						
26	Net weight	24-25						
27	Weight of barium	26x0.59						
28	Carrier added	-						
29	Chemical yield factor (Y)	28/27						
26	Net weight	Copy						
30	Thickness correction (T)	Graph						

Date _________________ Sample Number

		Operation						
31	Corrected sample (Bq)	23x29x30						
	STANDARD DEVIATION							
32	Background/count interval	4/3						
33	Sample/count intervals	21/20						
34	S^2	32+33						
35	S for sample (net cps)	√34						
36	Total correction factor	18x29x30						
37	S for sample (Bq)	35x36						