SSI-rapport 93-13

Statens strålskyddsinstitut
Swedish Radiation Protection Institute

Postadress
Box 60204
104 01 STOCKHOLM

Gatuadress
Karolinska sjukhuset
Solna

Telefon
08-729 71 00

Jorma Suomela

Method for determination of radon-222 in water by liquid scintillation counting

ISSN 0282-4434

Pris: 25kronor

Statens strålskyddsinstitut
Swedish Radiation Protection Institute

Nummer / Number: 93-13

ISSN: 0282-4434

Datum / Date of issue: 1993-06-28

Titelblad / Title page

Författare / Author:
Jorma Suomela

Avdelning / Division:

Division of Radiochemistry and Radioecology

Dokumentets titel / Title of the document:

Method for determination of radon - 222 in water by liquid scintillation counting

Sammanfattning / Abstract:

The procedure for the determination of radon-222 by liquid scintillation counting is quite specific for this radionuclide.
Radon-222 is extracted readily from the water sample by an organic scintillant. The decay products of radon-222 will remain in the water phase whilst radon-222 will be extracted into the organic phase. Before measurement the sample is stored for three hours until equilibrium is reached between radon-222 and its alpha emitting decay products. The alpha activity from radon-222 and its decay products is measured in a liquid scintillation counter.

Nyckelord (valda av författaren) / **Key words** (chosen by the author):

Radon-222, water, liquid scintillation counting

Antal sidor / Number of pages: 7

Division of Radiochemistry
and
Radioecology

Method for determination of radon-222 in water by liquid scintillation counting

according to
ISO/TC147/SC3/WG6/ Working document N14
revised by
Jorma Suomela

Content

DETERMINATION OF RADON-222 IN WATER BY LIQUID SCINTILLATION COUNTING

1. Principle

The procedure for the determination of radon-222 by liquid scintillation counting is quite specific for this radionuclide.

Radon-222 is extracted readily from the water sample by an organic scintillant. The decay products of radon-222 will remain in the water phase whilst radon-222 will be extracted into the organic phase. Before measurement the sample is stored for three hours until equilibrium is reached between radon-222 and its alpha emitting decay products. The alpha activity from radon-222 and its decay products is measured in a liquid scintillation counter.

2. Apparatus

Liquid scintillation counter

Low background liquid scintillation vials with a capacity of at least 20 ml.
The use of polyethylene scintillation vials is not recommended because of potential losses of radon through the walls of the vial.
The use of teflon or viton seals in the caps of the vials is recommended to ensure that there is no escape of radon from the vial.

Hypodermic syringe, disposable, 10-15 ml capacity.

3. Reagents

Distilled or deionized water, stored in a glass bottle for at least a month.

Scintillation solution. Commercially available toluene based scintillation cocktails should be satisfactory but should be tested for their suitability prior to routine use.

Certified Ra-226 standard solution.

4. Sampling

Prior to sampling liquid scintillation vials are prepared containing 10 ml of liquid scintillation solution.

If the water is supplied by a tap allow the water to flow freely for several minutes, without aeration, prior to sampling.

Carefully draw up a sample of 10 ml from the source of the water into a disposable syringe ensuring that there is no aeration of the sample. Special care is required when collecting samples to prevent significant loss of radon-222 by out-gassing.

When sampling surface water sources submerge the syringe well below the surface of the water and draw 10 ml of water into the syringe.

If it is desired to obtain a larger sample volume, or if it is not practical to transfer the sample to the scintillant vial at the site of sampling, a sample of water should be collected in a solid glass bottle of known volume which can be tightly capped after filling. The bottle should be filled slowly and smoothly from the source to minimise the loss of any radon from the sample. If possible allow the water to overflow from the bottle to ensure that it is completely filled and that no air gap will remain after sealing. Immediately re-cap the bottle. The sample should be transferred to the laboratory without delay for preparation prior to measurement.

5. Procedure

The sample in the hypodermic syringe is immediately, in situ, transferred to the liquid scintillation vial containing the scintillation solution. Inject the sample at the bottom of the vial, below the immiscible scintillation solution. Carefully withdraw the syringe needle from the vial and immediately secure the cap of the vial tightly. Shake the vial vigorously. Radon-222 will be extracted from the water phase to the organic scintillant solution due to its greater solubility in organic liquids, such as toluene. Note the time at which the collection of the sample takes place. At this stage sample vials can be transferred to the laboratory for measurement.

Counting procedure

The counting procedure should be carried out as soon as possible after returning samples to the laboratory. After shaking the counting vials, carefully wipe the outside of each vial with a cloth dampened with ethanol or similar solvent and leave them aside for a minimum of 3 hours to allow for ingrowth of the short-lived decay products of radon-222. Radon-222 and its short-lived daughters will be in equilibrium after 3 hours.The calibration of the counting system in the manner described will take into account any degree of disequilibria between radon-222 and its daughters in the organic phase and will be consistent with measurements on samples.

Place the counting vials in the liquid scintillation counter and count each vial for a preset period of time using a calibrated window for alpha counting. Ensure that, when transferring vials from storage to the counter after the 3 hour ingrowth, they are not shaken as this will greatly disturb the state of equilibrium between radon-222 and its short-lived daughters in the organic scintillant. Note the time and date at which counting commences. A suitable counting period will depend upon the activity concentration of Rn-222 in the sample but 5000 seconds per vial should usually be sufficient for low level measurement.

Determine the background of the counting system by counting a vial with 10 ml of the organic scintillant solution and 10 ml of deionized or distilled water. The background should be measured over the same spectral range as the samples and for at least the same counting period.

6. Determination of efficiency

Procedure

For the determination of efficiency of counting in the situation where the sample vail contains equal volumes of the water sample and the organic scintillant, accurately dilute a Ra-226 standard solution with distilled water and prepare a set of calibration solutions in liquid scintillation vials to cover the anticipated activity range of the samples to be analysed.

The total volume of aqueous solution in each vial should be 10 ml. Add 10 ml of the organic scintillant solution and tightly seal the cap of the vial. Leave these vials aside in a dark place

for at least 25 days to allow for secular equilibrium to be achieved between radium-226 and radon-222. At the end of the buildup period, shake the calibration vials and leave for a further period of 3 hours before counting. The radium-226 remains in the aqueous phase and does not contribute significantly to the count rate from the organic scintillant. Count each vial and calibrate the liquid scintillation counting systems as below.

Calculation of Efficiency

For a single calibration solution, the efficiency can be calculated using the expression:

$$E = \frac{R - R_0}{A_s} \qquad F(I)$$

where

- E is the counting efficiency for the system, in number of pulses per second counted per becquerel of radium-226 in the calibration solution
- R is the counting rate of the calibration solution (i.e. radon-222 and its decay products), in pulses per second;
- R_0 is the counting rate of the blank counting vial, in pulses per second and
- A_s is the activity of radium-226 in the calibration solution, in becquerel.

An alternative approach is to plot the measured count rate, for the set of calibration solutions, against the known radium-226 (i.e. radon-222) activity in each solution. The count rate varies linearly with radon-222 activity and, by linear regression of the data, determine the slope to yield the overall efficiency of the system, expressed as the number of pulses per second per becquerel of radon-222.

7. Calculations

7.1 Determination of radon-222 in the sample

To determine the activity concentration of Radon-222 in a water sample, use the following equation:

$$C_s = \frac{(R - R_0) \exp(\lambda \Delta t)}{V \times E} \qquad F(II)$$

where

- R it the gross counting rate for the sample, in pulses per second;
- R_0 is the counting rate for the blank counting vials, in pulses per second;

λ is the decay constant for Rn-222, in days $^{-1}$, i.e. 0.182 days $^{-1}$
V is the volume of the sample, in liters
Δt is the time interval, in days, between the collection of the sample and the commence of the counting;
E is the counting efficiency (see F (I))

7.2 Lower limit of detection

The estimate of the lower limit of detection, C_{min} becquerel per liter can be calculated according to the following equation:

$$C_{min} = \frac{2k\sqrt{\frac{R_o \quad (t + t_o)}{t \times t_o}}}{V \times E} \qquad F(III)$$

where

k is the confidence coefficient for type I errors. It is recommended to set k = 1.64, which is equivalent to 95% confidence level for the detection of activity;

t is the counting period for the sample, in seconds;

t_o is the counting period for the blank vial, in seconds; and

R_o and V are as defined in F(II) and

E is as defined in F (I)

With k = 1.64 and $t = t_o$ the the equation reduces to:

$$C_{min} = \frac{4.65\sqrt{\frac{R_o}{t}}}{V \times E}$$

As an example:

With R_o = 0.017 counts per second;
t = 10000s;
V = 0.01 liter
E = 2.90

then $C_{min} = \frac{4.65\sqrt{0.017 / 10000}}{0.01 \times 2.90} = 0.2$ Bq per liter

7.3 Standard deviation in the activity concentration

Calculate the standard deviation in the activity concentration of the sample due to the statistical nature of radioactive decay and background radiation from the following equation:

$$S_c = \frac{\sqrt{\left(\frac{R}{t}\right) + \left(\frac{R_o}{t_o}\right)}}{V \times E \exp(-\lambda \Delta t)} \qquad F(IV)$$

where

S_c is the standard deviation of C_s in becquerel per liter;

E is as defined in F (I)

R, R_o, λ, Δt and V are as defined in F (II); and

t and t_o are as defined in F (III)

8 Interference by other radionuclides

The presence of short-lived radium isotopes, Ra-223 and Ra-224 in the water sample may give rise to additional radon isotopes, respectively Rn-219 and Rn-220, through radioactive decay. It is possible that these radon isotopes will be extracted into the organic scintillant along with Rn-222. However, it is unlikely that either of these two radon isotopes will contribute significantly to the overall count rate of the sample. Rn-219 as well as Rn-220 are extremely short-lived isotopes compared to Rn-222 and will decay rapidly once they are extracted into the scintillant phase and no longer are supported by their parent radionuclides.

Other potential sources of interference would arise if there are relatively high levels of beta- or gamma emitting radionuclides in the water sample. Some of these nuclides might produce luminescence in the scintillant. Significant interference may occur with samples of water e.g. from mineral processing.

9. Bibliography

(1) Pritchard H.M. and Gesell T.F. "Rapid Measurements of Rn-222 Concentrations in Water with a Liquid Scintillation Counter', Health Physics 33 (1977), pp. 577-581.

(2) Pritchard, H.M., Gesell, T.F. and Meyer, C.R. 'Liquid Scintillation Analyses for Radium-226 and Radon-222 in Potable Waters', in 'Liquid Scintillation Counting, Recent Applications and Development', eidited by Chin-Tzu Peng, D.L. Horrocks and E.L. Alpen. Academic Press, (1980), New York, USA.

(3) Parks, N.J. and Tsuboi, K.K. 'Emulsion Scintillation Counting of Radium and Radon', Int. J. Appl. Rad. and Isotop. 29 (1978), pp. 77-80.

(4) Kimiko Horiuchi and Yukio Murakami. 'A New Procedure for the Determination of Radium in Water by Extraction of Radon and Application of Integral Counting with a Liquid Scintillation Counter', Int. J. Appl. Rad. and Isotop. 32 (1981), pp. 291-294.

(5) Graves B. (Ed.). 'Radon, Radium and Other Radionuclides in Ground Water - Hydrogeologic Impact and Application to Indoor Airborne Contamination', Proceedings of the NWWA Conference April 7-9, 1987, Somerset, New Jersey, Lewis Publishers, Michigan, 1988.

(6) Cothern C.R. and Smith J.E., (Eds.). 'Environmental Radon', Environmental Science Research Vol. 35, Plenum Press, New York, 1987.

(7) Currie, L.A. 'Limits for Qualitative Detection and Quantitative Determination - Application to Radiochemistry', Anal. Chem. 40 (1968), pp. 586-593.

(8) PArsa B. and Horton T. 'Radon-222 in Drinking Water: An NJDEP-EERF Collaborative Study', Health Phys. 58 (1990), pp. 209-212.

(9) Vitz E. 'Toward a Standard Method for Determining Waterborne Radon', Health Phys. 60 (1991), pp 817-829.

(10) Lowry J.D. 'Measuring Low Radon Levels in Drinking Water Supplies', J. Amer. Water Works Assoc. 83 (1991), pp. 149-153.

(11) ISO/TC147/SC3/WG6/Working Document N14.

Utgivna SSI-rapporter 1993

SSI-reports published in 1993

Nummer Number	Titel Title	Författare Author
01	Publikationer	Informationsenheten
02	Survey and Evaluation of the External Research and Development Programme 1977-1983 of the Swedish Radiation Protection Institute	Lars Persson
03	SSIs granskning av SKBs forskningsprogram 1992	Mikael Jensen
04	Ett arbetsnormalinstrument för kontinuerlig radon-mätning	Hans Möre
05	LENA_P A probabilistic version of the LENA code version 1.0 January 1993	Ulf Bäverstam Olof Karlberg
06	Studies on the Waste Depository of the Sillamäe Plant State of the art report from the project group	Hans Ehdwall et al
07	Medical Uses of Radiation: Retaining the Benefit but Recognising the Harm Presented at the OECD/NEA Workshop on Radiation Protection Toward the Turn of the Century, Paris, 11-13 January 1993	Jack Valentin and Geoff Webb
08	Projekt Strålskydd Öst Lägesrapport	Torkel Bennerstedt,
09	Isotopkommittérapporter 1991	Mauricio Alvarez
10	Radon 1993 En rapport över läget	Statens strålskyddsinstitut
11	Methods for determination of strontium-90 in food and environmental samples by Cerenkov counting	Lena Wallberg J. Suomela, J. Melin
12	Method for determination of radium-226 in water by liquid scintillation counting	Jorma Suomela
13	Method for determination of radon-222 in water by liquid scintillation counting	Jorma Suomela