AUTO-RADON: A NEW AUTOMATIC LIQUID SCINTILLATION SYSTEM FOR MONITORING RADON IN WATER AND AIR

G Jónsson • P Theodórsson[1] • K Sigurdsson

Science Institute, University of Iceland, Dunhaga 3, IS-107 Reykjavík, Iceland.

ABSTRACT. A simple, robust single-phototube liquid scintillation counting system for continuously monitoring ^{222}Rn (radon) in water was developed at the Science Institute and has operated for 1 yr. It can also measure radon in air. A 20-mL open vial with 15 mL of scintillator is above the flat end of a 28-mm-diameter, vertical phototube. Water runs slowly through a 40-cm-long silicone tube wound in a spiral above the vial. Radon diffuses through its wall and is absorbed in the scintillator until equilibrium concentration between the 2 liquids is reached. For monitoring radon in air, air is bubbled slowly through the scintillator. Only ^{214}Po (half-life 0.16 μs) α pulses, identified through pulse-time analysis, are recorded in the Rn register. The results of 6-hr counting periods are stored on a diskette. For an unshielded detector, the Rn background is 3 counts per hour and ^{214}Po counting efficiency 91%. With 15 mL of scintillator, a ^{214}Po count rate of 3 times the standard deviation of the background (in 6-hr periods) corresponds to about 1 mBq/L in water and 1 Bq/m^3 in air.

INTRODUCTION

During the past 2 decades, radon has been monitored extensively in geophysical research and environmental studies, generally through discrete sampling and measuring the samples in a laboratory. In Iceland, Rn in groundwater was studied from 1977 to 1992 as a possible earthquake precursor (Hauksson and Goddard 1981; Jónsson and Einarsson 1996), using a Lucas cell for measuring Rn. These studies were resumed in 1999 with greatly improved measuring techniques, using liquid scintillation counting (Gudjonsson and Theodórsson 2002). Water samples (200 mL) were collected twice a week at 6 geothermal boreholes in a seismically active area in south Iceland, sent to our institute, and measured in an automatic single photomultiplier tube (PMT) liquid scintillation (LS) counter made in our laboratory (Theodórsson and Gudjonsson 2000).

For a continuous record of Rn concentration and to save earlier work, we began in 2001 the development, based on our experience with the laboratory-operated system, of a relatively simple single-PMT LS system for automatic operation in geothermal well pumping stations. The performance of the prototype (Gudjonsson and Theodórsson 2002) was not satisfactory then, but the system, Auto-Radon, has been greatly improved via a number of steps and works now to our full satisfaction. It has been in operation for a year. This system replaced the earlier laboratory Rn measurements in December 2004.

METHODS

Detector System and Radon Transfer from Water to Scintillator

An important feature of the new system is the simple and reliable technique for transferring Rn from water to the scintillator. This transfer caused many problems in the early phases of the work. We tested various solutions; first, Rn was transferred by a stream of air bubbled through the water and then the scintillator. This was later replaced by a process where air was circulated and bubbled through the 2 liquids in a closed system by a small peristaltic pump. To our surprise, we discovered that Rn was lost by diffusion through the wall of the silicone tube used. Although this is a known phenomenon, the rapidity of the diffusion came as a surprise to us. We turned this to our advantage as described below.

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[1] Corresponding author. Email: pth@raunvis.hi.is.

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Figure 1 shows schematically the detector unit used today, which is inside an aluminium tube. A vertical PMT (Hamamatsu R6094) faces the bottom of a conventional 22-mL vial with 5–15 mL of mineral oil scintillator, but is separated by a 3-mm-thick acryl plate glued to the tube, to make the upper Rn compartment air-tight. The PMT is 28 mm in diameter and the length of its bulb is 87 mm. For a more compact detector unit, we also tested a shorter PMT (Hamamastu R1924A) of 25 mm diameter with a 43-mm-long bulb. Its counting performance is practically the same as for R6094, but the tube is somewhat more expensive. Figure 2 shows the Rn pulse-height spectrum.

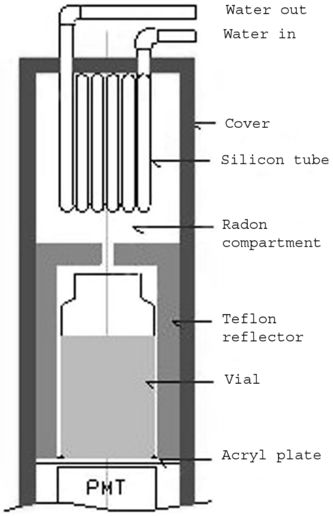

Figure 1 Scheme of the detector unit

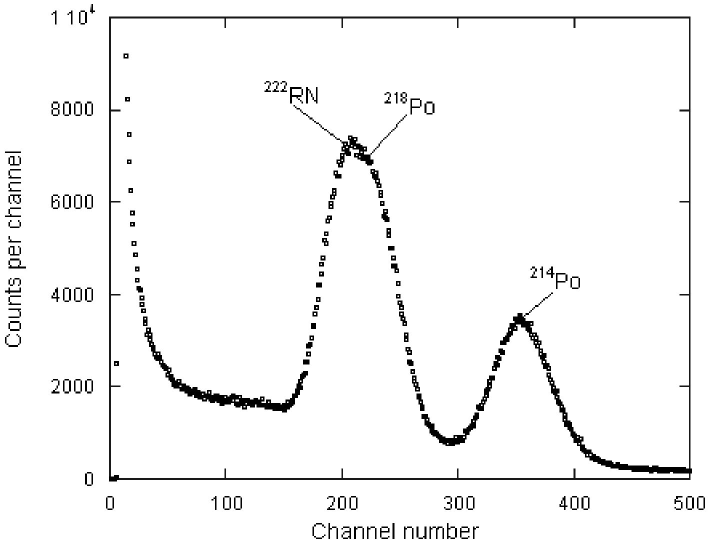

Figure 2 Spectrum of ^{222}Rn and its short-lived decay products for a mineral oil scintillator.

The water to be monitored runs slowly (~30 mL/min) through a 40-cm-long silicon tube spiral above the open vial. The outer diameter of the tube is 5 mm with a 1-mm-thick wall. Rn diffuses reversibly through the wall and is absorbed in the scintillator until an equilibrium concentration is reached, determined by the relative solubility of Rn in the 2 liquids. At steady-state, the Rn concentration in the scintillator is 52× higher than in water (Bem et al. 1994). This method of Rn transfer eliminated the peristaltic pump used in an earlier version, which caused problems.

Radioactive secular equilibrium is established in the scintillator after ~2 hr in the chain starting with ^{222}Rn and ending in ^{214}Po, where 3 α particles and 2 β particles are emitted (Table 1). The Rn response time is, however, dominated by the slow diffusion of Rn in the scintillator, as shown below.

Table 1 The short-lived ^{222}Rn series.

Nuclide	Half-life	Particle, energy (MeV)
^{222}Rn Rn	3.82 d	α 5.49
^{218}Po RaA	3.05 min	α 6.00
^{214}Pb RaB	26.8 min	β 0.65
^{214}Bi RaC	19.7 min	β 3.17 (23%), 1.75 (77%)
^{214}Po RaC'	0.16 ms	α 7.69
^{210}Pb RaD	22 yr	β 0.018

Electronic Unit and ^{214}Po Counting

A specially designed, laboratory-made electronic unit delivers high voltage to the PMT, amplifies its anode pulses, and its voltage discriminators sort them into 4 pulse-height windows. A laptop computer, connected online to the electronic unit, receives the signal from the 4 discriminators, processes it, and stores the results in internal memory.

Using pulse-time analysis, the computer separates and records the α pulses of the short-lived ^{214}Po (half-life 0.16 µs), 96% of which come within 1 µs after the pulse of its beta-emitting mother nuclide, ^{214}Pb (Table 1). This specific Rn counting mode reduces the background of the unshielded detector to ~3 pulses/hr and provides good protection from external disturbances (Theodórsson and Gudjonsson 2000). The ^{214}Po detection efficiency is 91%.

The number of Rn pulses accumulated in 6-hr periods is stored on a diskette, replaced with a new one every second week, and the removed diskette is posted to the Science Institute.

A compact microprocessorized electronic unit, needing no computer except to start the counting, is being developed. It will send the results once a day by SMS message to a central computer.

SYSTEM PERFORMANCE

To study how fast the system responds to Rn concentration changes in water, it was operated in a geothermal borehole station close to our laboratory with 5 mL of scintillator. First, the background count rate was recorded for 40 hr with a radon-free scintillator in the vial (Figure 3). Then, the stream of borehole water was turned on and Rn was measured for 3 d; it reached an equilibrium concentration in ~1 d. Assuming a constant Rn concentration in the borehole water during this period, the rise curve can be described by:

$$A(t) = A_f\,(1-e^{-t/\tau}) \tag{1}$$

where $A(t)$ is the count rate at time t; A_f is the final (equilibrium) count rate; and τ is the response time, the time needed to reach 63% equilibrium concentration. The response time is mainly deter-

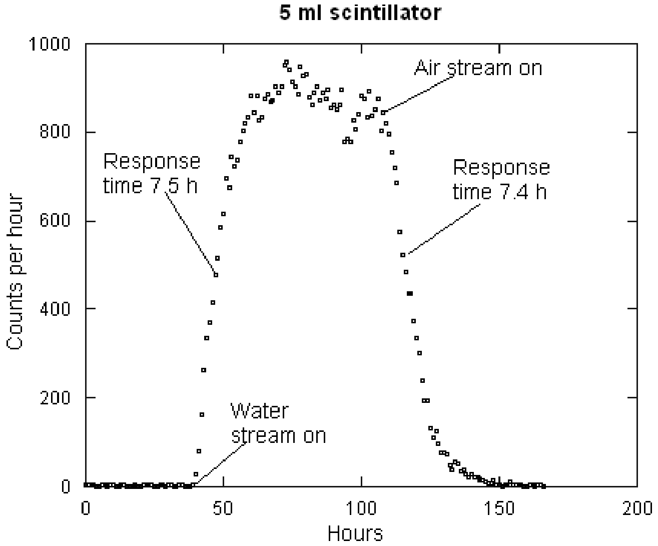

Figure 3 Test runs in a geothermal well pumping station to study response time

mined by the slow diffusion of Rn in the scintillator, and is 7.4 hr with 5 mL of scintillator. In our Rn monitoring work, however, we use vials with 15 mL of scintillator to increase the sensitivity.

About 4.5 d after the start of the experiment, air was pumped through a silicon tube at 20 mL/min. The stream of air flushes out the Rn in water with practically the same response time, as expected.

Figure 4 shows a 35-d Rn record at one of the measuring stations. The sensitivity increases with the volume of scintillator, but the response time increases at the same time. With 10 mL of scintillator, the response time is ~10 hr, and 19 hr for 15 mL. Since a 19-hr response time is quite acceptable in our case, we have selected this volume for our routine work.

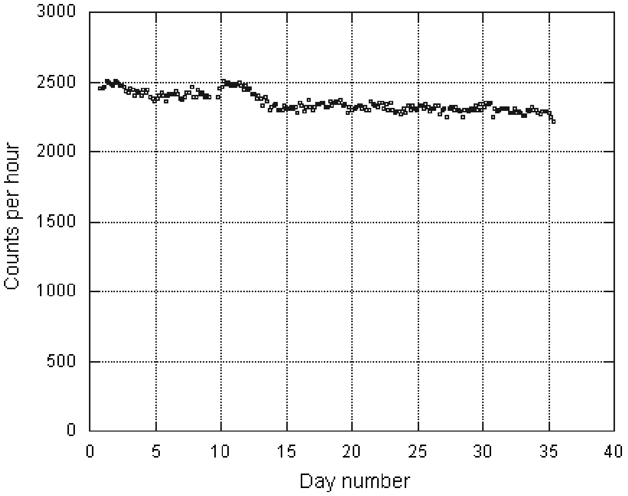

Figure 4 Rn record at one of the monitoring stations

The system was calibrated by measuring the Rn concentration in the water that was being monitored with the LS counting method of Prichard and Gesell (1977): 6 samples of 10 mL of water to which 10 mL of scintillator is added. This resulted in a 1.33 Bq/L Rn concentration in the water. By transforming the count rate of Auto-Radon to concentration by using the 91% counting efficiency and calculating that the concentration in the scintillator (15 mL) is 52× higher at equilibrium than in the water, we get 0.98 Bq/L. This result is acceptable, but the calibration must be studied further.

CONCLUSION

A simple, reliable LSC system for measuring Rn in water and air was built and tested. Rn is transferred from water to the scintillator by free diffusion through the wall of a silicon tube. This eliminated the mechanical pump used for air circulation in our earlier model. Only ^{214}Po (half-life 0.16 online μs) α pulses, identified through pulse-time analysis, are recorded in the Rn register. With 15 mL of scintillator, the Rn background for an unshielded detector is 3 counts per hour and the ^{214}Po counting efficiency is 91%. The background count rate corresponds to a water Rn concentration of ~1 mBq/L.

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REFERENCES

Ben H, Bakir YYY, Bou-Rabee F. 1994. An improved method for low-level ^{222}Rn determination on environmental waters by liquid scintillation counting with pulse shape analysis. *Journal of Radioanalytical Chemistry Letters* 186(2):119–27.

Gudjonsson GI, Theodórsson P. 2000. A compact automatic low-level liquid scintillation system for radon-in water measurements by pulse pair counting. *Applied Radiation and Isotopes* 53(1–2):377–80.

Gudjonsson GI, Theodórsson P. 2002. A simple and sensitive liquid scintillation counting system for continuous monitoring of radon in water. In: Möbius S, Noakes JE, Schönhofer F, editors. *LSC 2001, Advances in Liquid Scintillation Spectrometry.* Tucson: Radiocarbon. p 249–52.

Hauksson E, Goddard JG. 1981. Radon earthquake precursor studies in Iceland. *Journal of Geophysical Research* 86(B8):7037–54.

Jónsson S, Einarsson P. 1996. Radon anomalies and earthquakes in south Iceland seismic zone 1977–1993. In: Thorkelsson B, editor. *Seismology in Europe*. Reykjavík: European Seismological Commission. p 247–52.

Prichard HM, Gesell TF. 1977. Rapid measurements of ^{222}Rn concentrations in water with a commercial liquid scintillation counter. *Health Physics* 33:577–81.

Theodórsson P, Gudjonsson GI. 2003. Increased radon detection sensitivity: extraction from 200 mL of water and liquid scintillation counting. *Health Physics* 85: 610–2.