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Building and characterisation of a radon emanation source using a historical watch painted with radioluminescent paint containing radium

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Abstract

Conducting experiments with ^{222}Rn not always requires a radon emanation source traceable to primary standards, often it is sufficient to use a well characterised radium source. In this work we present the building and characterisation of a radon emanation source using a watch with radioluminescent paint containing ^{226}Ra. The source with 179 ± 9 kBq ^{226}Ra was characterised for different air flow rates between 0.05 L/min and 2 L/min. The mean radon emanation coefficient was 0.54 ± 0.19 while a dependence of radon emanation on relative air humidity was discovered.

1 Introduction

Radon is a natural radioactive noble gas which is the second leading cause for lung cancer after smoking [1]. Measurements of and experiments with radon are still of great interest leading to a need of controllable radon sources independent of local circumstances such as geology. The production of radon emanation sources traceable to primary standards are often described and require large efforts [2, 3, 4]. In applications where no traceability to primary standards is needed alternative methods to produce a radon emanation source are available.

One possibility is the usage of natural ore and rock. Another option is the usage of ^{226}Ra in form of radioluminescent paint. Throughout the 20th century some dials on clocks, watches and the indicators on instruments and gauges for vehicles were painted with radioluminescent paint to make them readable in the dark without the need for electric light [5, 6, 7]. Until today these instruments can be found in use for example in military applications such as vehicles of all kinds. Also these dials are subject of collectors and museums.

The decay of ^{226}Ra results in gaseous ^{222}Rn which is able to emanate from the luminescent paint and leave the instrument through leakages. This can lead to increased radon gas concentrations in places where even only small quantities of such instruments are stored. In this paper we took advantage of the wide availability of such gauges in Switzerland and the resulting radon output and present the production and characterisation of a flow-through radon emanation source using a watch with radioluminescent paint containing ^{226}Ra.

2 Materials and Methods

2.1 Radon emanation

Let A_0 be the activity of ^{226}Ra in the source, let $\lambda_{Rn} = 2.1 \times 10^{-6}\,\mathrm{s}^{-1}$ be the decay constant of ^{222}Rn and let f be the air flow through the radon emanation source. Then the radon activity concentration in the outgoing air is given by

$$a_{Rn} = \chi \lambda_{Rn} \frac{A_0}{f} \tag{1}$$

where χ is the emanation coefficient of the source.

With the known value of λ_{Rn} and measurements of A_0, f and a_{Rn}, the emanation coefficient of the source can be calculated as

$$\chi = \frac{a_{Rn} f}{\lambda_{Rn} A_0}. \tag{2}$$

2.2 Radium dial and steel container

As described above we used a watch containing a dial painted with radioactive luminescent paint with ^{226}Ra. The watch is shown in figure 1a. The flange at the back side of the watch was removed prior to the experiments. The activity of ^{226}Ra was measured with a high purity germanium detector to be $179 \pm 9\,\mathrm{kBq}$.

As a holder for the source a stainless steel container was built, it is shown in figure 1b. The interior of the container is cylindrical with length 100 mm and diameter 72 mm therefore the inner volume is $4.1 \times 10^5\,\mathrm{mm}^3$. Both ends of the

(a) Image of the watch used in this experiment. The numbers and pointers are painted with radioluminescent paint containing ^{226}Ra. The diameter of the dial is around 60 mm. *Picture: Labor Spiez, Switzerland.*

(b) The stainless steel container used to host the watch. On the top of the container the two valves can be seen. The removable plate to access the interior is located on the bottom of the container.

Figure 1: The two components for the radon emanation source. On the left side the watch with radioluminescent paint, on the right side the stainless steel container.

cylinder are closed with steel plates, on one side the two valves are screwed into two holes connecting to the inside. On the opposite side the steel plate can be removed to access the interior of the container. The source, i.e. the watch, was placed inside the steel container through the access on the bottom side with the dial facing up towards the valves.

2.3 Measurement and characterisation

First the source was characterised in terms of radon emanation for different air flow rates. Next the emanation coefficient χ was determined and finally the dependence of radon emanation on environmental parameters was investigated. All measurements of radon activity concentration and environmental parameters were done with Bertin Instruments AlphaGUARD of type DF2000. The device was also recently calibrated at the Federal Institute of Metrology METAS in Switzerland prior to any measurements.

Before starting any measurements with the source we measured the radon activity concentration in the laboratory room and that of the empty stainless steel container to assure that no previous contamination is present neither in the container nor in the laboratory itself. The measurement in the laboratory room was running for 24 h, the measurement device was placed in the same location where further experiments were conducted. For the measurement with the empty steel container both valves were opened and one valve was connected to the inlet of the measurement device with an air hose the other valve was open to the laboratory air. The internal pump of the device was set to 2 L/min, this measurement was done over 1 h. After these measurements the watch was placed inside the stainless steel container. In the following we refer to the steel container with the watch inside as the radon emanation source or simply the source.

For the characterisation of the source we used the same setup as with the empty steel container, where one valve is directly connected to the measurement device and the other valve is opened to the laboratory air. Additionally the outlet air from the measurement device was directly connected to an exhaust duct from the laboratory via a plastic tube to prevent high radon concentrations accumulating in the laboratory room. Then the internal pump of the measurement device was set to different air flow rates namely 0.05 L/min, 0.1 L/min, 0.2 L/min, 0.3 L/min, 0.4 L/min and 0.5 L/min. For each air flow rate a measurement of radon activity concentration over 50 min to 70 min was done. Furthermore a measurement with an air flow rate of 1 L/min was done over 130 min. To verify the stability of the radon emanation we conducted a last measurement with an air flow rate of 2 L/min over 89 h. From these measurements we later determined all characteristics of the radon emanation source.

3 Results

The radon activity concentration in the laboratory room was measured to be $10\pm4\,\mathrm{Bq/m^3}$. Measuring the empty container resulted in $5\pm2\,\mathrm{Bq/m^3}$. The detailed results of the radon emanation measurements for different air flow rates are shown in figure 2 and summarised in table 1. The stability of the radon emanation at 2 L/min was assessed over 89 h and the mean radon emanation was $3616\,\mathrm{Bq/m^3}$ with maximum of $6747\,\mathrm{Bq/m^3}$ and minimum $2570\,\mathrm{Bq/m^3}$, the standard deviation was $643\,\mathrm{Bq/m^3}$. The radon emanation coefficient from equation **??** is evaluated for each air flow rate and also shown in table 1. The average emanation coefficient over all air flow rates was 0.54 ± 0.19. Furthermore in figure 3 the pressure, relative humidity and temperature are shown against the radon emanation coefficient for the air flow of 2 L/min.

4 Discussion

The laboratory background and the steel container without the radium source inside were measured resulting in $10\pm4\,\mathrm{Bq/m^3}$ and $5\pm2\,\mathrm{Bq/m^3}$ respectively. These values are very low compared to the average radon concentration in occupied

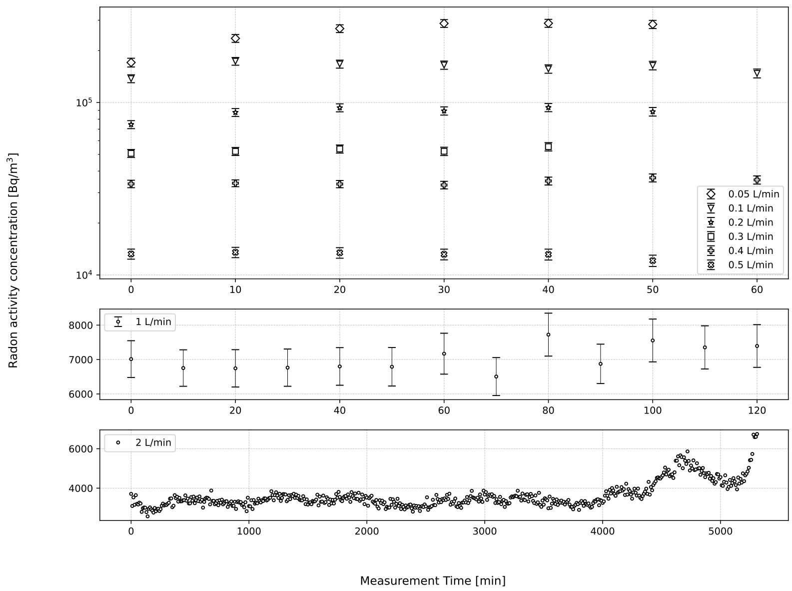

Figure 2: Detailed results for the radon emanation measurement for different air flow rates. In the upper chart air flow rates between 0.05 L/min and 0.5 L/min are shown. In the second chart the results of the measurement with 1 L/min and in the bottom chart the results of the measurement with 2 L/min are shown. Each measurement point represents the integration over 10 min, the errors shown were given by the measurement device. For the measurement with 2 L/min errorbars are omitted for better readability.

Air flow [L/min]	^{222}Rn [Bq/m^3]	rel. humidity [%]	χ [1]
0.05	$255\,544 \pm 42\,129$	57.1 ± 0.9	0.57 ± 0.10
0.10	$158\,670 \pm 11\,524$	58.9 ± 0.1	0.70 ± 0.06
0.20	$87\,790 \pm 6356$	61.9 ± 1.1	0.78 ± 0.07
0.30	$52\,823 \pm 1634$	61.8 ± 0.1	0.70 ± 0.04
0.40	$34\,558 \pm 1132$	60.7 ± 0.4	0.61 ± 0.04
0.50	$13\,108 \pm 467$	40.5 ± 1.2	0.29 ± 0.02
1.00	7033 ± 357	43.3 ± 0.3	0.31 ± 0.02
2.00	3616 ± 643	42.9 ± 5.0	0.32 ± 0.06

Table 1: Table showing the mean values for the radon activity concentration of the outgoing air and radon emanation coefficient for different air flow rates. On the left side is the air flow rate which was set on the radon measurement device's pump, in the second column are the mean values of the radon activity concentration with standard deviation in Becquerels per cubic meter. The third column shows the measured relative humidities. On the right side the calculated radon emanation coefficients for each air flow rate are given.

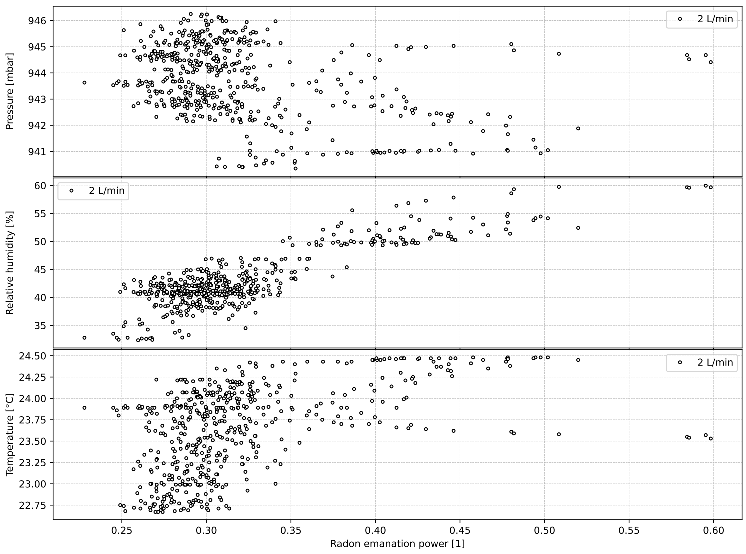

Figure 3: The pressure, relative humidity and temperature shown against radon emanation coefficient at 2 L/min. For better readability errorbars are omitted.

indoor spaces in Switzerland of 75 Bq/m^3 [8]. Therefore it was shown, that there is no elevated radon concentration in the laboratory and no previous contamination with radium in the steel container.

Next the results of the characterisation of the source are shown in figure 2. For 0.3 L/min, 0.4 L/min and 0.5 L/min the radon activity concentration was randomly fluctuating but generally stable over the measurement duration. For 0.05 L/min and 0.2 L/min there was an increase of radon activity concentration over the first few measurement points, i.e. the first few tens of minutes, this is also slightly the case for 0.1 L/min. This might indicate that some time is needed for the radon emanation to reach a steady state after changing the air flow rate. This effect seems to be larger for low air flow rates around 0.1 L/min than for 0.5 L/min. The standard deviation of the mean radon emanation at 0.05 L/min was 42 129 Bq/m^3 which is 16 %, for 0.5 L/min the standard deviation was 467 Bq/m^3 which is only 4 %. A longer measurement duration for each air flow would compensate for such effects. The measurement with 1 L/min was conducted over 130 min and showed stable results. The mean value was 7033$\pm$357 Bq/m^3 which is a standard deviation of 5 %.

We also observed a dependence between radon emanation coefficient and relative humidity. In table 1 it is shown that for the first five measurements relative humidities around 60 % were measured and radon emanation coefficient between

0.57 and 0.78 were calculated. The last three measurements showed relative humidities around 40 % and radon emanation coefficients between 0.29 and 0.32. For the air flow of 2 L/min the detailed results of pressure, relative humidity and temperature versus radon emanation coefficient are shown in figure 3, the chart indicates that an increase in relative humidity leads to an increase in radon emanation coefficient. [9] generally investigated radon emanation coefficients for porous media, our findings are in alignment with what was found by the authors. For the pressure versus radon emanation coefficient no correlation can be seen, other so in the graphic for relative humidity versus radon emanation coefficient. For the temperature one could assume a slight correlation but there are many outliers in this dataset. It is important to notice that these three parameters are not completely independent. Since the laboratory is not air-conditioned the parameters are mainly determined by the weather.

The measurement with 2 L/min was running over 89 h to also investigate the long term stability of the radon emanation. As shown in table 1 the mean value was $3616 \pm 643\,\mathrm{Bq/m^3}$ which is a standard deviation of 18 %. In figure 2 the results over the whole duration are shown. Between minutes zero and 4000 small fluctuations can be seen, after around 4200 min the radon emanation strongly increased up to almost $6000\,\mathrm{Bq/m^3}$ and after a short decrease it rose again to around $6800\,\mathrm{Bq/m^3}$ at the end of the measurement. This was due to a significant increase in relative humidity. Before minute 4000 the relative humidity was between 32 % and 47 %, starting around 4200 min the relative humidity rose to 60 %. Further showing the correlation between the two properties.

5 Conclusion

In this experiment we built a radon emanation source from a historical watch containing radioactive luminescent paint with $179 \pm 9\,\mathrm{kBq}$ ^{226}Ra. The watch was placed inside a stainless steel container with valves and connectors for air pipes. The source was characterised in terms of radon emanation for different air flow rates and long-term stability. The mean radon emanation coefficient for air flow rates between 0.05 L/min and 2 L/min was 0.54 ± 0.19, over a long term measurement of 89 h at 2 L/min the mean radon emanation coefficient was 0.32 with a standard deviation of 0.06. The large standard deviations for all measurements where due to a dependence of radon emanation on relative humidity, which we did not control in this experiment. Therefore the radon emanation source does not fully satisfy the request for long term stability. Further experiments could more closely investigate the dependence of radon emanation on relative humidity. Additionally the radon emanation for a larger range of air flow rates could be investigated.

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