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A simple and versatile ²²²Rn/²²⁰Rn exposure chamber

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Abstract

A simple ²²²Rn/²²⁰Rn exposure chamber is described. The chamber is made of acrylic and has a volume of 0.41 m³. The ²²²Rn and ²²⁰Rn sources can be operated independently or simultaneously. The concentrations of ²²²Rn and ²²⁰Rn were monitored by a continuous alpha spectrometer called RAD7. The capability of alpha spectrometry has removed the mutual interference between ²²⁰Rn and ²²²Rn, which is a major problem in measuring ²²⁰Rn. Furthermore, the capability can also provide fast response to changes in the ²²²Rn level and ²²⁰Rn level in the exposure chamber, with temporal resolutions of about 20 and 2 min, respectively. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

It is established that the tracheobronchial deposition of ²²²Rn (radon) progeny in the human body can lead to lung cancers, and people have become more and more aware of the ²²²Rn problem nowadays. There are various methods to measure concentrations of ²²²Rn and its progeny, and the reliability of these methods invariably depends on the validity and accuracy of the calibrations involved. Exposure chambers are almost indispensable for calibration of instruments for measuring ²²²Rn and its progeny. Besides, exposure chambers are often designed in such a way that other characteristics of ²²²Rn

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progeny can also be studied, such as the potential alpha energy concentration (PAEC) of the progeny and the size distribution of the PAEC.

It has been proposed that the problem of ²²⁰Rn (thoron) should not be overlooked [1]. In recent years, there is increasing attention to the problem. Research and survey on the behavior of ²²⁰Rn and its progeny have been carried out [2-4] and have shown that the concentrations of ²²⁰Rn and its progeny are not negligible compared to those of ²²²Rn and its progeny. While calibrations using ²²²Rn exposure chambers are rather well established, the construction of exposure chambers of ²²⁰Rn or more importantly of mixed ²²²Rn/²²⁰Rn, and the associated problems are less studied. Many factors can make accurate measurements of ²²⁰Rn concentrations difficult. The first one is the presence of and interference from ²²²Rn. The second is the short half-life of ²²⁰Rn. As can be

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seen below, while the short half-life of ²²⁰Rn has simplified some aspects of its measurement, the sampling method is a critical issue.

More recently, Monte Carlo simulations have shown that the sensitivities of CR39 and LR115 detectors, which are the two commonly used solid state nuclear track detectors (SSNTDs), to ²²⁰Rn are at least comparable to those to 222 Rn. The errors introduced in measurements of ²²²Rn concentrations by the presence of ²²⁰Rn depend on a number of factors such as the ratio of ²²⁰Rn/²²²Rn concentrations and dimensions of the diffusion chamber [5.6]. Since SSNTD method is one of the most commonly employed to measure ²²²Rn concentrations, it is essential to analyze and quantify the influence of ²²⁰Rn through experimental studies. An exposure chamber of mixed ²²²Rn/²²⁰Rn is in need for this purpose. The current paper describes the construction of such an exposure chamber. Sample exposure conditions achieved by the exposure chamber will also be presented.

2. The ²²²Rn/²²⁰Rn exposure chamber

The exposure chamber was made of acrylic with a size of $1.1 \times 0.61 \times 0.61$ m³ (=0.41 m³). The size of the chamber is designed to be small to allow changes in the 222 Rn and/or 220 Rn levels inside the chamber on relatively small time scales. A block diagram of the chamber is shown in Fig. 1. The air flow pattern into and out of the exposure chamber

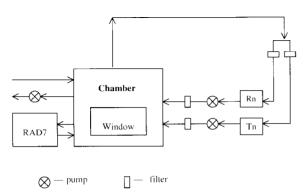


Fig. 1. Block diagram of the ²²²Rn/²²⁰Rn chamber.

is determined by different pumps and valve controls. The chamber is mainly operated in a recirculation mode, but operation in the flow-through mode is also possible. At the center of the front panel is a door with a size of $0.40 \times 0.36 \, \text{m}^2$ (=0.144 m²) which can be opened for putting various equipment or detectors into the exposure chamber.

2.1. 222Rn and 220Rn gas supply

The ²²²Rn source is ²²⁶Ra while the ²²⁰Rn source is ²²⁸Th (both purchased from Isotope Products Laboratories, California). Both sources had an activity of 10 uCi (370 kBa) as of 17 August 2000. The sources were formed as evaporated metallic salts in ceramic matrix which were affixed to the internal bottom of 10 ml vials. Air is passed through the vial but not through the source to avoid getting the radionuclides out of the ceramic matrix. The gas from each source is driven by an individual diaphragm pump (GAST POA-P101-BN. from Gast Manufacturing, Inc., MI) through a membrane filter (pore size of 0.45 µm) into the chamber. In the recirculation mode, the output air from the chamber is recirculated through the ²²²Rn and/or ²²⁰Rn sources. The free air flow rate of each diaphragm pump is 1.6 m³/h. The ²²²Rn and ²²⁰Rn sources can be operated independently or simultaneously.

2.2. ²²²Rn and ²²⁰Rn monitor—RAD7

The instrument RAD7 (from Durridge Company Inc., MA) forms the basis of a comprehensive ²²²Rn and ²²⁰Rn monitor for our exposure chamber. Air from within the chamber is drawn to the RAD7 inlet first through a small drying tube filled with dessicant and then through a 0.45 µm filter, and the air from the outlet of RAD7 is then recirculated into the chamber. The filter prevents ²²²Rn and ²²⁰Rn progeny from getting into RAD7, so that RAD7 can measure the ²²²Rn and ²²⁰Rn gas concentrations alone. The pump of the RAD7 will start running when the preset program is executed. The pump is microprocessor controlled and the flow rate is around 650 ml/min.

There are different continuous ²²²Rn/²²⁰Rn monitors on the market, and most of these are based on detection of alpha particles. There are currently three types of alpha particle detectors being used as electronic ²²²Rn/²²⁰Rn monitors, namely, scintillation cells (or "Lucas" cells), ion chambers and solid state alpha detectors, RAD7 has an internal sample cell of a 0.71 hemisphere, with a solid state ion-implanted planar silicon alpha detector at the center. The inside of the hemisphere is coated with an electrical conductor which is charged to a potential of 2-5 kV relative to the detector, so that positively charged progeny decayed from ²²²Rn and ²²⁰Rn are driven by the electric field towards the detector. When a progeny atom reaches the detector and subsequently decays and emits an alpha particle, the alpha particle has a 50% probability of being detected by the detector. As a result, an electrical signal is generated with the strength being proportional to the alpha energy. RAD7 will then amplify and sort the signals according to their energies [7].

The RAD7 spectrum is a scale of alpha energies from 0 to 10 MeV, which is divided into 200 channels each of 0.05 MeV width. The alpha energies associated with ²²²Rn and ²²⁰Rn are in the range of 6–9 MeV. These 200 channels are grouped into eight energy windows (labeled as A–H). Windows A–D are the functional ones.

Window A: records 6.00 MeV alpha particles from ²¹⁸Po, which has a half-life of 3 min, and is thus the window for ²²²Rn sniff mode or "new" ²²²Rn.

Window B: records 6.78 MeV alpha particles from 216 Po, which has a half-life of 0.15 s, and is thus the window for 220 Rn sniff mode or "new" 220 Rn.

Window C: records 7.69 MeV alpha particles from ²¹⁴Po, which has a half-life of nearly an hour, and is thus the window for "old" ²²²Rn.

Window D: records 8.78 MeV alpha particles from ²¹²Po, which has a half-life of about 10 h, and is thus the window for "old" ²²⁰Rn. For every 66 counts in window D, there should be 34 counts (6.05 and 6.09 MeV) in window A due to the two-way split from ²¹²Bi, so the counts in window A

are automatically corrected for counts in window D by RAD7.

RAD7 adds windows E-H together to form window O (for "other"), which is mainly for diagnostic purposes.

RAD7 allows different modes of measurements, such as the normal mode and the sniff mode. For our purposes, the sniff mode is employed. In the sniff mode, RAD7 uses only ²¹⁸Po signals to determine the ²²²Rn concentrations and ²¹⁶Po signals to determine the ²²⁰Rn concentrations, ignoring subsequent and long-lived progeny.

Our RAD7 instrument was calibrated by the manufacturer on 20 February 2001. The 222 Rn sensitivity in the sniff mode is 0.233 CPM/(pCi/l), and the 220 Rn sensitivity in the sniff mode is 0.117 CPM/(pCi/l). The 220 Rn calculations is based on a standard setup calibrated by the manufacturer. A small drying tube (6 in.) filled with 30 g of Drierite dessicant, three feet of vinyl tubing with an inner diameter of $\frac{3}{6}$ in, and the male Luer type inlet filter (pore size of 0.4–1.0 µm) are all standardized.

2.3. RAD7 settings for monitoring ²²²Rn and ²²⁰Rn in the exposure chamber

For ²²²Rn measurements, in the sniff mode, the RAD7 counts only ²¹⁸Po decays. Since ²¹⁸Po has a half-life of 3.05 min, the ²¹⁸Po atoms on the detector surface takes about five half-lives, or around 15 min, to reach equilibrium with the ²²²Rn gas in the chamber. Cycle times of 10 min were adopted, which would provide a response time to variations in ²²²Rn concentrations with a time scale of about 20 min.

For ²²⁰Rn measurements, the main problem arises from the decay during its acquisition since ²²⁰Rn itself has a half-life of less than a minute. The ²¹⁶Po, whose decays we count in the sniff mode, has a half-life of only 150 ms, so the response of RAD7 to ²²⁰Rn, once the ²²⁰Rn is in the chamber, is instantaneous. For most of our experiments, the RAD7 is also kept on 10-min cycles, and it will print out a ²²⁰Rn value as well as a ²²²Rn value at the end of every cycle.

2.4. Computer interface

The RAD7 has an RS232 serial port that can be used for transfer of data into a file on a personal computer. A computer software (Capture 1.2.0) is provided by the manufacturer for this purpose. Besides this, for ²²²Rn/²²⁰Rn concentrations which vary on small time scales, a small and simple program can be written to perform the time integration of ²²²Rn/²²⁰Rn concentrations to evaluate the exposure conditions (in units of Bq h m ³), which is useful for calibration of detectors

Furthermore, although not being employed at present for our exposure chamber, the ²²²Rn/²²⁰Rn concentrations in the exposure chamber can also be controlled by RAD7 with the optional RADLINK software and an external relay box. Two thresholds can be set for each of the two relays in the box. At the end of each measurement cycle, RAD7 will compare the measured ²²²Rn/²²⁰Rn concentrations with the values set for each of the thresholds and turn on or off the relays accordingly. In this case, no extra external controller is required.

3. Advantages of the present ²²²Rn/²²⁰Rn exposure chamber

The greatest advantage of RAD7 we exploit for monitoring ²²²Rn and ²²⁰Rn levels in our chamber is its capability of alpha spectrometry, i.e., the energy of each particle can be electrically determined. This gives detailed information on the ²²²Rn or ²²⁰Rn progeny being detected (e.g., ²¹⁸Po, ²¹⁴Po, etc.), from which the amount of new or old ²²²Rn, or ²²⁰Rn can be individually determined.

Therefore, one of the difficulties encountered in measuring ²²⁰Rn levels, namely the interference from ²²²Rn, can be comfortably removed by RAD7. RAD7 separates ²²²Rn and ²²⁰Rn signals and counts them at the same time with no interference between them.

Another great advantage associated with the capability of alpha spectrometry is the fast response to changes in the ²²²Rn and ²²⁰Rn level in the exposure chamber. When an instrument

draws in air with ²²²Rn and makes measurements. the ²²²Rn will also decay within the instrument and there should be buildups of ²²²Rn progeny. The relatively long-lived ²²²Rn progeny, ²¹⁴Po, takes many hours to decay away. The RAD7 is capable of focussing at ²¹⁸Po alone and discarding information on ²¹⁴Po, and since ²¹⁸Po has a halflife of only 3 min. RAD7 has a response time of only 15 min to changes in ²²²Rn levels. Regarding ²²⁰Rn, on the contrary, ²¹⁶Po has a half-life of 150 ms, so RAD7 has an instantaneous response to changes in ²²⁰Rn levels effectively. As mentioned earlier, we employed the sniff mode for monitoring our exposure chamber, for which RAD7 uses only ²¹⁸Po signals to determine the ²²²Rn concentrations and ²¹⁶Po signals to determine the ²²⁰Rn concentrations. In this way, RAD7 has a fast response to changes in the ²⁵²Rn and ²²⁰Rn levels.

4. Results and discussion

Sample exposure conditions achieved by the exposure chamber are presented in this section. In Figs. 2 and 3, the curves for buildup and clearance of ²²²Rn alone and ²²⁰Rn alone, respectively, in the ²²²Rn/²²⁰Rn chamber are shown. The ²²²Rn gas or ²²⁰Rn gas was pumped into the chamber in a recirculation mode for 60 min, and clearance was started at 60 min. Both the data for the 222Rn or ²²⁰Rn concentrations were reported at the end of every 10 min. The maximum achievable ²²²Rn or ²²⁰Rn concentrations are both around 140 kBgm⁻³. From Figs. 2 and 3, owing to the fast response of RAD7, we can observe the variations of the ²²²Rn or ²²⁰Rn concentrations on a relatively small time scale. The capability of generating these fast variations of the ²²²Rn or ²²⁰Rn concentrations in the exposure chamber, together with the ability to quantify such changes, may be useful to test the response of some detectors, such as the characteristics of a diffusion barrier for the activated charcoal canister [8-10] and the optimization of the skim-off technique using the charcoal canister method [11,12]. Moreover, the fast response can also provide more accurate time integration of the ²²²Rn and ²²⁰Rn concentrations to evaluate the exposure conditions (in units of Bghm³).

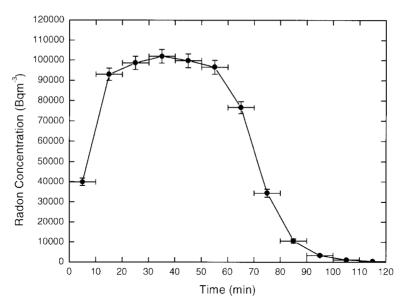


Fig. 2. The curve for buildup and clearance of 222 Rn alone in the 222 Rn/ 220 Rn chamber. 222 Rn was pumped into the chamber in a recirculation mode for 60 min, and clearance was started at 60 min. Data for the 222 Rn concentrations were reported at the end of every 10 min

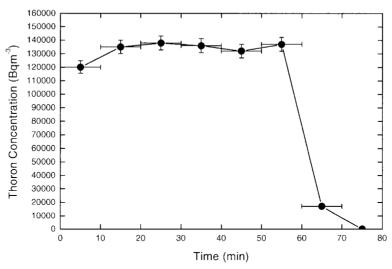


Fig. 3. The curve for buildup and clearance of 220 Rn alone in the 222 Rn/ 220 Rn chamber. 220 Rn was pumped into the chamber in a recirculation mode for 60 min, and clearance was started at 60 min. Data for the 220 Rn concentrations were reported at the end of every 10 min.

Fig. 4 shows the curves for buildup and clearance of ^{222}Rn and ^{220}Rn together in the $^{222}Rn/^{220}Rn$ chamber. Here, ^{222}Rn and ^{220}Rn were pumped into the chamber simultaneously in a

recirculation mode for 60 min, and clearance was started at 60 min. The data for the ²²²Rn and ²²⁰Rn concentrations were again reported at the end of every 10 min. From this figure, it is obvious

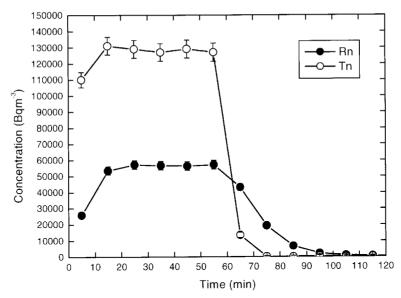


Fig. 4. The curves for buildup and clearance of 222 Rn and 220 Rn together in the 222 Rn/ 220 Rn chamber. 222 Rn and 220 Rn were pumped into the chamber simultaneously in a recirculation mode for 60 min, and clearance was started at 60 min. Data for the 222 Rn and 220 Rn concentrations were reported at the end of every 10 min.

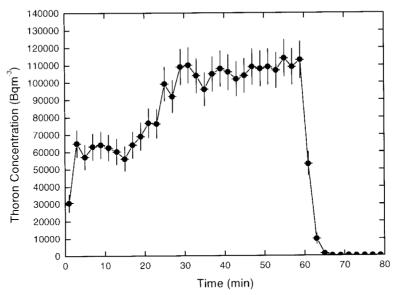


Fig. 5. The curve for buildup and clearance of 220 Rn alone in the 222 Rn/ 220 Rn chamber. 220 Rn was pumped into the chamber in a recirculation mode for 60 min, and clearance was started at 60 min. Data for the 220 Rn concentrations were reported at the end of every 2 min.

that the signals from ²²²Rn and ²²⁰Rn inside the exposure chamber are separated and counted at the same time with no mutual interference. This

directly demonstrates the removal of the difficulty introduced by the interference from ^{222}Rn in measuring ^{220}Rn levels.

As mentioned before, RAD7 has an instantaneous response to changes in ²²⁰Rn levels effectively, since ²¹⁶Po has a half-life of 150 ms. Therefore if only exposure to ²²⁰Rn is required. a much shorter cycle time of 2 min can be adopted. An example is shown in Fig. 5. Here, a curve for buildup and clearance of ²²⁰Rn alone in the ²²²Rn/²²⁰Rn chamber is shown, where ²²⁰Rn was pumped into the chamber in a recirculation mode for 60 min and clearance was started at 60 min. The data for the ²²⁰Rn concentrations were reported at the end of every 2 min. It can be seen that variations in the 220Rn concentrations with a much smaller time scale can be recorded. In this way, a far more accurate time integration of the ²²⁰Rn concentrations can be performed to evaluate the exposure conditions (in units of Bahm 3).

In the above, we have presented the characteristics of a simple and versatile ²²²Rn/²²⁰Rn exposure chamber. Although the current exposure chamber is not designed for studies on the characteristics of ²²²Rn and ²²⁰Rn progeny such as the size distribution of the potential alpha energy concentration (PAEC), inclusions of these measurements only represent one further step on top of the current design. Note that the methodologies for determinations of the PAEC of ²²²Rn and ²²⁰Rn progeny and the size

distribution of PAEC are more or less well established.

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