"FUNGI"- A RADON MEASURING INSTRUMENT WITH FAST RESPONSE

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Abstract

The FUNGI uses an electrostatic field to collect the daughter products of radon decay, and a solid state detector to resolve the energies of radium A (^{218}Po) and radium C' (^{214}Po) alpha particles. This permits a great degree of flexibility in the measurement of radon and radon daughters. In addition, by limiting the instrument response to the radium A channel only, the response to changes in radon concentration of this instrument is ten times faster than other instruments, making it suitable for radon survey applications.

Introduction

One of the problems encountered in radon survey applications is the long half life of radium C (^{214}Bi), which effectively contaminates the detector for several hours before it decays. This is particularly true when the ratio of the maximum radon concentration to the minimum radon concentration is high. To overcome this, most instruments use a filter paper to collect the radon daughters and then transport the filter to the detector where it is counted. Such instruments are rather complex because they require a pump to force the air through the filter paper and some mechanical means to transport the filter to the counting station. In addition, the counting after sampling method that these instruments use is five times less sensitive than the counting during sampling method used by the FUNGI.

Previous work with radon measuring instruments using an electrostatic field for the collection of radon daughters has been reported. Costa-Ribeiro, et al¹ have described an instrument which uses foam to remove radon daughters from air as it diffuses into the 28 cm³ detection volume. An electrostatic field collects radon daughter products produced in this volume by the decay of radon gas. An alpha sensitive photographic plate is used to record the alpha emissions. M. E. Wrenn, et al² have designed an instrument with a volume increased to one liter and with an alpha scintillator counter to record alpha emissions. In this report, the design of a ten liter instrument which uses a solid state alpha detector is described. This permits RaA (^{218}Po) and RaC' (^{214}Po) alphas to be resolved separately. By detecting only the short half life RaA daughters, significant improvement in the response to changing levels of radon concentration has been achieved. The principle components of the FUNGI are: an outer shell of open-pore foam which permits the diffusion of air containing radon, but filters out radon daughters, a decay volume in which radon daughter products are formed, and a detector. The daughter products in the decay volume are collected on the detector by an electrostatic field. The detector registers the alpha emissions from the collected daughter products.

Theoretical

The operation of the instrument can be compared with the two filter method as developed by Thomas and LeClare.³ The radon concentration equation for their method is:

$$C_{\text{Rn}} = \frac{X}{Z} \cdot \frac{0.45}{EV F_f} \quad (1)$$

where C_{Rn} is the radon concentration in picocuries per liter; X is the number of counts observed during the counting period; E is the counting efficiency; V is the decay volume in liters; and F_f is the collection efficiency of the radon daughters generated in the decay volume (some daughters are lost on the surfaces of the decay volume). Z is a factor which relates alpha disintegrations (X/E) during the counting period to the rate of radium-A atoms arriving at the second filter during the sampling period.

The relationship given in equation (1) can be used for the instruments described in this report (the second filter is replaced with a detector), but the Z factor, which holds for counting after sampling, must be replaced with a new factor, Z', for counting during sampling. This involves the integration of the activity equations for radium-A and radium-C'. It can be shown⁴ that

$$Z' = Z'_A + Z'_C \quad (2a)$$

$$Z'_A = (T_2 - T_1) + 4.4 (e^{-\lambda_1 T_2} - e^{-\lambda_1 T_1}) \quad (2b)$$

$$Z'_C = (T_2 - T_1) + 0.103 (e^{-\lambda_1 T_2} - e^{-\lambda_1 T_1}) + 164.68 (e^{-\lambda_2 T_2} - e^{-\lambda_2 T_1}) - 93.30 (e^{-\lambda_3 T_2} - e^{-\lambda_3 T_1}) \quad (2c)$$

where Z'_A and Z'_C relate the disintegrations of radium-A and radium-C', respectively; T_1 and T_2 are the beginning and end of the measurement interval in minutes; and λ_1 , λ_2 , and λ_3 are the decay constants of radium-A, B, and C.* After a few hours (approximately 2.5), as can be seen from equation (2), the Z' factor reduces to $2(T_2 - T_1)$ or twice the measurement interval (minutes). For example, in a constant radon concentration, the

* $\lambda_1 = 0.22726$
 $\lambda_2 = 0.25864$
 $\lambda_3 = 0.035185$

steady-state Z' for a 30 minute measurement is 60.

A considerable improvement in sensitivity results with the counting during sampling method. This can be shown by comparing the two factors Z and Z' for the same measurement time; in a two-hour measurement, the maximum Z is 34 (one-hour sample, one-hour count), whereas Z' from 0-2 hours is 170. Comparisons for other measurement intervals show that counting during sampling is more sensitive than counting after sampling by a factor of five or more.

Instruments With Scintillation Detectors

Fig. 1 is a photograph of the first experimental instrument. Since it resembles a large mushroom, it, and subsequent instruments, has been tagged with the name FUNGI.

Fig. 1. 3.75 Liter FUNGI

The unit consists of a 3.75 liter hemisphere covered with 2.5 cm of open-pore foam. The open-pore foam permits the diffusion of the radon gas, but filters out radon daughters.¹ Filter paper can be used instead of the open-pore foam, but radon diffuses faster through the foam. A wire mesh screen supports the foam and is the anode for the electrostatic collection of charged radon daughters.

The scintillation detector is located at the center of the hemisphere, Fig. 2. A layer of aluminized mylar covers the scintillator to form a cathode. In operation, it was found that the photomultiplier tube gave erratic results when the hemisphere was at ground potential and negative voltage was applied to the aluminized mylar. This was eliminated by grounding the aluminized mylar and applying positive voltage to the hemisphere. No collection was observed with negative potential on the hemisphere; this can be seen from Fig. 3. Thus, there are no negatively charged daughters, a finding that is in agreement with Wexler.⁵

"FUNGI" SCINTILLATION UNIT

Fig. 2. FUNGI Cross-Section

Fig. 3. Collection vs. Voltage. The curve shows that there is no collection with negative potentials and, hence, no negatively charged radon daughter products.

The FUNGI was tested by placing it in a known radon concentration of approximately 30 pCi/l and then calculating F_f by using equation (1) with Z' substituted for Z . Ideally, F_f should be 100%, but the initial series of tests indicated a value of only 20%. This low value was traced to the base plate of the hemisphere which was initially plastic. When this plastic was covered with aluminum foil that was electrically connected to the hemisphere screen, F_f more than doubled from 20% to 50%. Hence, to obtain maximum collection, voltage gradients across insulators must be electrically screened. In an attempt to increase F_f above 50%, much higher electric fields were studied by increasing the voltage and reducing the size of the hemisphere. Even with 20kV applied to the smaller hemisphere, the value of F_f remained at 50-60%. From this, it was concluded that

recombination is not the primary cause for the low value of F_f .

The transient response of the FUNGI was measured by subjecting it to a step change in radon concentration. The three curves in Fig. 4 show the results.

Fig. 4. Transient response to step change in radon concentration. A is raw data, B and C are calculated results which show that response is less than 30 min.

Curve A is the raw counting data recorded in each 30 minute measurement interval; curve B is the ratio of the counts measured during a 30 minute interval to the corresponding Z' for that interval. For example, in the interval 120-150 minutes, Z' is obtained by setting $T_1 = 120$, $T_2 = 150$, and using equation (2). Curve C is the ratio of the total counts recorded to the corresponding Z' . In this case, Z' is obtained by setting $T_1 = 0$ and $T_2 = 30, 60, 90$, etc. Curves B and C are almost identical and reach a steady value within the first 30 minute measurement period. This indicates that the transient response of the FUNGI to a step change in radon concentration is less than 30 minutes.

Humidity Effects

While testing the FUNGI, it was observed that F_f was significantly lower in the summer than it was in the winter. This prompted a series of tests to determine the effects of humidity.

Humidity tests were conducted with two instruments--the 3.75 liter FUNGI and a larger 10 liter unit having cylindrical geometry. Both units exhibit a linear humidity dependence with the same slope, namely a 6% decrease in sensitivity for a 10% increase in relative humidity. Fig. 5 is a plot of the data for the 3.75 liter FUNGI. Since the data for the humidity tests was taken at a constant temperature of 27°C., the abscissa in Fig. 5 is directly proportional to the water content of the air.

Another humidity effect observed was the failure of the unit to achieve its original sensitivity after being exposed to high humidity. The original

Fig. 5. FUNGI Response vs. Relative Humidity

sensitivity was restored only after placing the instrument in a vacuum chamber; the cause of this hysteresis is believed to be water retention in the foam.

One solution to the humidity problem is to dry the air before it enters the decay volume by placing a layer of activated silica gel desiccant* over the foam. This method has been tested and is very effective. However, the crystals have to be replaced (or oven dried) approximately every two weeks, depending on the humidity. In addition, the radon gas diffuses much more slowly through these crystals than the open-pore foam (3-4 hours versus less than 0.5 hr.). Therefore, applications requiring a rapid measurement of radon require pumping for a few minutes until the air in the instrument's decay volume is representative of the air being measured.

Limited temperature testing of these instruments indicated no temperature dependence from 21°C. to 38°C.

Solid State Detector

Since the FUNGI type of instrument uses electrostatic collection which deposits the radon daughters right on the detector surface, experiments were begun to see if a solid state detector might achieve reasonable resolution of the radium-A and radium-C' energies. Initial experiments with a conventional solid state detector gave excellent energy resolution, but the collection efficiency, F_f , was very poor (less than 10%). This low value of F_f is due to the metal case, Fig. 6, of the conventional detector; most of the radon daughters are attracted to this metal case, and the alpha radiation from these radon daughters is not measured. Fig. 7 illustrates the initial attempt to correct this. Here, the detector is

* TEL-TALE - Davison Chemical, Baltimore, Md.

DETECTOR CONSTRUCTION

Fig. 6. Solid state alpha detector construction. Conventional metal case detector is shown at top and printed circuit ring mount detector at center and bottom.

- NOTE: 1. POLYCARBONATE FILM TM. "KIMFOL"
KIMBERLY - CLARK CORP.
2. TEFLON TAPE PLACED ON DETECTOR TO
PREVENT BREAKDOWN ON WIRE CONNECTION
TO THE GOLD EVAPORATION

Fig. 7. Method used to focus charged daughters to active area of metal case detectors.

covered with a 1.9 micron thick polycarbonate film which has a circular area of 100 µg/cm² gold deposited on it. By keeping the gold at a negative potential (-500V), most of the radon daughters are attracted to the gold surface which has the active area of the detector beneath it. While this scheme

works, the resolution suffers in passing through the air space between the polycarbonate film and the detector surface.

At the present time, a special detector manufactured to our specifications is being used.* This detector has an active area of 300 mm² and uses a printed circuit ring mount, as shown in Fig. 6, instead of a metal case. Electrical connection to the detector surface is made via a small conducting tab on the printed circuit ring mount. To determine the collection loss caused by the mount, tests were done with filter paper placed on the detector and using the conventional two-filter method, equation (1). Filter papers of two diameters were used; one covered only the active area of the detector, while the other was larger and extended to the end of the ring mount. By measuring the collection on the filter papers, it was determined that 95% of the collection takes place on the detector active area and 5% is lost on the printed circuit ring mount.

Instruments With Solid State Detectors

Tests were conducted with the solid state detector mounted in the center of one end of a 10 liter cylinder and with a mesh screen covered with open-pore foam at the other end. The cylinder walls are solid aluminum and are operated with a potential of +1.5kV with respect to the detector.

Figs. 8 and 9 show the resolution of the detector with and without drying the air entering the decay volume. In both cases, the resolution (FWHM) is an

Fig. 8. Alpha spectrum with dry air

excellent 37 keV. However, there is a marked difference in the ratio of the two peaks for dry air and humid air; with dry air, the ratio of the counts in the radium-C' region to the counts in the radium-A region is 1.2 to 1, while with humid air, the ratio is 2:1. The effect of moisture is to lower the counts in both peak regions, but the radium-A peak is affected much more than the radium-C' peak. With dry air, the overall efficiency, F_f is 58%; the efficiency for

*Princeton Gamma-Tech, Princeton, New Jersey

Fig. 9. Alpha spectrum with humid air

radium-C' is 63%; and the efficiency for radium-A is 52%. With moisture, the F_f is only 20%, while the efficiencies for radium-C' and radium-A drop to 27% and 13.5%, respectively.

Since each radium-C' atom is produced from a radium-A atom, the lower collection efficiency of radium-A indicates a loss mechanism, probably recombination. Correcting for this loss, the greatest F_f (dry air) becomes 63%. To this, the 5% detector loss is added, which yields a corrected F_f of 66%.

Charge Experiments

Having established that there are no negatively charged radon daughters, Fig. 3, one interpretation of the 66% F_f is that some (34%) of the radon daughters are uncharged. To explore this further, the apparatus shown in Fig. 10 was used to perform charge experiments.

Fig. 10. Apparatus to determine charge of radon daughter products.

Basically, it consists of a small (.503 liter) two-filter radon chamber with a wire along the axis. A section in front of the first filter is used to dry or add moisture to the air entering the chamber.

With the wire at ground potential, several runs were taken with dry air at a flow rate of 5 liters/minute. After sampling the air, the second filter is moved to a counting station, and equation (1) is used with the appropriate Z to determine F_f . The zero voltage F_f is 68%, which is in good agreement with the theoretical F_f of 62%.¹ With +5kV applied to the wire, F_f drops to 21%. Therefore, $\frac{.21}{.68}$ or 31% of the radon daughters are uncharged and 69% are charged. The 69% agrees very well with the maximum F_f previously determined (66%).

When the above tests were repeated with moisture-laden air, the values of F_f , with and without voltage, were the same as for dry air. Further testing indicated that the saturation voltage is 100V for dry air and 700V for humid air. Thus, the decrease in sensitivity caused by humidity is due to recombination. By scaling the electric field corresponding to 700V, a one liter hemisphere would require 4,300V and a ten liter hemisphere would require 22kV to overcome humidity-induced recombination; this assumes that only the magnitude of the electric field need be considered.

Tests at higher radon concentrations are now being done. Initial results at 550 pCi/l indicate that 80% of the daughters are charged and 20% uncharged. Additional tests with the FUNGI are needed to confirm these higher concentration results.

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