CURRENT STATE OF THE ART IN MEASURING ENVIRONMENTAL RADON

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Abstract

According to the US EPA, radon is the leading cause of lung cancer among non-smokers and the second leading cause overall. Data from several epidemiological studies show definitive evidence of the association between indoor radon exposure and lung cancer.
In the past 30 years, emphasis has been on measuring radon rather than radon progeny concentrations because of the simplicity, convenience and cost effectiveness of radon measuring instruments and methods. Using an equilibrium ratio between radon and radon progeny of 0.4-0.5, radon concentration measurements can be converted to working level and to exposure in working level month. In recent years, over 1 million short-term measurements for radon were made annually using grab sampling, integrating and continuous radon devices. As a result of these short-term measurements more than 800,000 residences with elevated radon levels were identified and were mitigated successfully. This paper will emphasize the current development of different instruments and methods, their sensitivities, practicality and cost effectiveness for making short-term measurements of environmental radon. More than 99% of indoor measurements involve radon only. Radon progeny, used mostly in research and diagnostics, will not be discussed.

Introduction

According to the US Environmental Protection Agency (EPA) and the World Health Organization (WHO) Handbook on Indoor Radon (WHO, 2009) radon is the second leading cause of lung cancer after smoking. It is estimated that of the 160,000 annual lung cancers in the US, about 20,000 are due to inhalation of radon and radon progeny. Data reported from several epidemiological studies indicate that exposure to indoor radon causes lung cancer.
In light of the latest scientific data on radon epidemiology, WHO proposed a reference level or action level of 2.7 pCi/L to minimize the health hazard due to indoor radon exposure. In the US alone, more than 20 million measurements of radon were made in the last 25 years and about 800,000 residential buildings were mitigated successfully. In its national survey, EPA estimated that one in every fifteen homes in the US or about 7% have radon concentration levels greater than the EPA action level of 4 pCi/L. More recent measurement data by radon professionals and mitigators, suggest that one in every 10 homes may have radon concentration levels above the EPA action level.

The increased interest in measuring radon in recent years has stimulated instrument research and development to meet the demand by radon measurement professionals, home inspectors, mitigators, real estate professionals, environmental firms, researchers and the general public.

Today, emphasis is on radon measurements rather than on radon progeny because radon measurements are simpler, more convenient, and more practical, providing a good estimate of the radiation exposure to the general public in a timely fashion. If we assume that about 90% of the radon measurements are <4 pCi/L, it is very important that the measurement devices have adequate sensitivity to measure environmental radon levels. Since more than 95% of the radon measurements in the US are short-term lasting 2-7 days, and more frequently 2-3 days, to accommodate the needs of real estate transactions, a measuring device with adequate sensitivity should be selected to ensure accurate radon measurements. In addition, all measurement methods should incorporate quality assurance and quality control measures to assure the reliability of radon measurements.

The instruments discussed in this paper are primarily based on the experience of the authors and from instrument brochures, published information and from the instrument evaluation process conducted by the U.S. EPA, the National Radon Safety (NRSB) and the National Environment Health Association – National Radon Proficiency Program (NEHA-NRPP).

Instrument Measurement Methods and Techniques

The measurement techniques for radon utilize the alpha and gamma ray radiations emitted by radon and its decay products. The most commonly used instruments for characterizing the indoor radon environment are passive integrating devices and continuous electronic monitors used mostly for short-term measurements.

In the U.S., long-term alpha track detectors and long-term electret ion chambers are used to confirm the performance of a mitigation system on an annual basis or for yearly determination of outdoor radon. In Europe, Canada, Asia and other countries long-term radon measurements lasting from 90-365 days are emphasized over short-term measurements. Instruments and methods for grab sampling are hardly used in the US any longer. The scintillation cell for grab sampling was ideal for making measurements in underground mines, in research, and to a small extent, in continuous scintillation cell radon monitors early on in the Radon Metrology Program. Figure 1 shows a 1 liter scintillation cell coupled to a 5-inch photomultiplier tube.

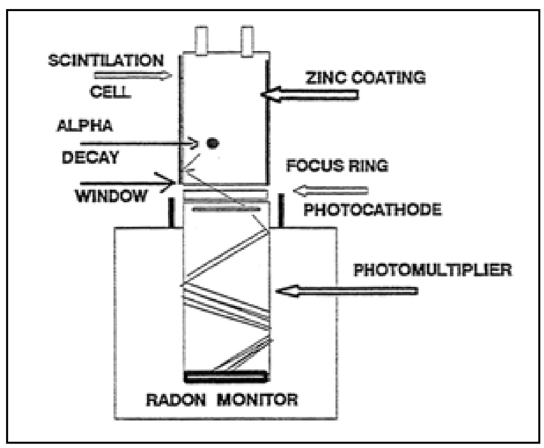

Figure 1. Scintillation cell with PMT

Activated Carbon Collectors (AC)

Activated carbon collectors in metal or plastic containers or bags containing (25-90 g) of activated carbon are the only passive devices that utilize gamma ray counting from the decay of the radon progeny Pb-214 and Bi-214 that are in equilibrium with radon inside the sealed container. Figure 2 shows the Airchek and the RTCA charcoal collectors. Activated carbon collectors of the open-face type are used typically for 2-4 day exposures and diffusion barrier collectors that minimize the amount of water vapor competing for the carbon active sites can be used from 2-7 day exposures. Diffusion barrier collectors provide a very accurate average radon concentration even if the radon during the test varies by more than a factor of 10 and if humidity is high because they adsorb less water vapor and minimize the effect of strong air movement. (George and Weber 1990, U.S. EPA 1992 and George 1996). The performance of activated carbon collectors depends heavily on the QA and QC procedures used during preparation (such as determining batch to batch charcoal variation) and system calibration under different conditions of temperature, humidity and duration of exposure. Calibration factors should be determined for different conditions encountered in the field. Testing laboratories that follow and exercise these steps provide accurate results.

Figure 2. Airchek (top); RTCA charcoal canister (bottom)

The lower level of detection (LLD) for activated carbon collectors depending on the type and size, ranges from 0.1 - 0.2 pCi/L of radon in a 4-day test. Activated carbon collectors are the most sensitive short-term radon devices with tight standard deviation of duplicates. Radon is continuously being adsorbed and accumulates on the carbon decreasing by decay while in other instruments such as continuous radon monitors the radon is detected while passing through the sensitive volume. The sensitivities of some activated carbon collectors are listed in Table 1.

Activated carbon collectors have become the most popular passive devices for measuring environmental radon (George, 1984, George, 1990, Cohen, 1986, Gray, 1990). They provide the average radon concentration during the duration of the exposure. Because some charcoal devices can be recycled and be used for numerous radon measurements they have become cost effective for testing laboratories.

Liquid Scintillation Vial (LS)

An alternate method, Figure 3, for measuring environmental radon is the LS vial method used by several laboratories. The RTCA 20 cc LS vial uses 2 g of activated carbon provided with a diffusion barrier to minimize the effect of humidity (George, 1997). Liquid scintillation collectors that contain 2-3 g of activated carbon in a 20 cc vial utilize the alpha radiation from Rn-222, Po-218 and Po-214. Some labs may count both the alpha and beta radiation from the radon progeny. The sensitivity

of the method listed in Table 1, is adequate to obtain accurate results for 2-7 day exposures. Like the charcoal canister, the

Figure 3. RTCA LS vial

LS vial provides the average radon concentration during the measurement period. Unlike the charcoal canister method, LS vials cannot be recycled after analysis.

About 70% of the short-term radon measurements in the U.S. are made with passive, open-face or diffusion barrier activated carbon collectors, and LS vials.

Electret Ion Chambers (EIC)

In electret ion chambers the electret is charged to a known voltage. When air enters the chamber, the alpha particles from Rn-222 and Po-218 and Po-214 ionize the air and discharge the voltage on the electret, which is measured with a voltage meter. Figure 4, shows the Rad Elec ionization chamber in the test position. The positively charged electret collects the negative air ions and becomes discharged according to the concentration of radon that gets inside chamber. The electret surface voltage is measured with a special voltage reader (Kotrappa, 1988). The voltage decreases in proportion to the concentration of radon that enters the chamber. The electret ion chamber measures the average radon concentration of a 2-14 day test.

Figure 4. Rad Elec electret ion chamber

Primary users of short-term electrets that read the electrets in the field are considered a mobile laboratory and are required to incorporate Standard Operation and QA and QC procedures. For short-term tests thick electrets can be used for multiple tests in the same ionization chamber until the final voltage is about 200 volts.
The biggest disadvantage of short-term electrets is their response to natural beta, gamma and X-ray radiation at the test site which can be a significant interference at radon concentration levels < 2-4 pCi/L. The manufacturer of the short-term electrets device makes a correction for the contribution of natural radiation in radon equivalent obtained from charts listing the radon equivalent at different altitudes where the tests are being conducted.

Figure 5. RTCA Radome electrets

To avoid this type of correction for the background contribution in long-term measurements, RTCA uses two side by side electret ion chambers as shown in Figure 5. One measures the contribution of radon and the background radiation and the second one measures directly the contribution of the background radiation only.

The more popular electret ion chamber used for short-term measurements (2-7 days) utilizes a thick electret yielding a voltage drop of 2.0 Volts/pCi/L Day. In the long-term electret ion chambers used for 90-365 days, a thin and less sensitive electret is used yielding about 0.15-0.17

Volts/pCi/L Day. The sensitivity of the two types of electret ion chambers is listed in Table 2. About 10% of the short-term radon measurements in the U.S. are conducted with electret ion chambers.

Alpha Track Detectors (ATD)

Alpha track detectors were used in some radon surveys conducted by EPA and in some States in the late 1980’s. They are more appropriate for the determination of the annual average concentration of radon. Figure 6, shows three types of alpha track detectors used in the US. (Accustar, Landauer, and RSSI). Today, they are infrequently used because of the real estate driven demand for short-term measurements. About 1-2% of the radon measurements in the U.S. use long-term alpha track detectors. We will describe them briefly to demonstrate what the rest of the world outside the US is using in their radon programs. In the US, some radon professionals or home-owners may use long-term alpha detectors to monitor mitigated homes on an annual basis.

Figure 6. Alpha track detectors. Top-bottom: Accustar, Landauer, RSSI

The principle of detection is based on the production of tracks made by the interaction of alpha particles in solid-state materials such as Cr-39 and LR-115 cellulose nitrate (Alter, 1981, Urban, 1981, and Vasudenan, 1994). After exposure, the tracks are made visible by chemical or electrochemical etching in a caustic solution. The tracks are counted by automated optical scanning systems or by image analysis. Accuracy depends on the area of plastic counted and on the concentration of radon. The number of tracks is proportional to the radon concentration as determined from calibration exposure tests.
Because the method is insensitive, exposures from 90-365 days are required to accumulate a sufficient number of tracks for good statistical counting.

Open face (bare) alpha track detectors that utilize the LR-115 plastic detectors are more sensitive at high altitudes. An increase in sensitivity was observed when exposed at an altitude of 5,000 feet (in the presence of less dense air). The sensitivities of different types of alpha track detectors using CR-39 plastic material listed in Table 3 and obtained from the literature and from personal communication range from 3.0 – 23.0 tracks/cm^2 per 4 pCi/L Day. Alpha track detectors require exceptional QA and QC procedures to characterize plastic variation from batch to batch and acquired background and temperature variation during production of the plastic detectors. One of the main disadvantages of alpha track detectors is the possibility that ambient radon can leak into the device if poorly sealed during shipping, handling and storage and produce unwanted alpha tracks that will overestimate the radon concentration.

Continuous Radon Monitors (CR)

In the past 25 years, several continuous radon monitors were developed that provide the real time radon measurements. They provide the hourly measurements and the average radon concentration during 2-3 days of testing.

Most of the commercial continuous radon monitors are passive where air diffuses in the sensitive volume passively and two are active where the radon is brought to sensitive volume of the instrument by means of an air pump. They are electronic computerized instruments. They are useful in real estate transactions where results are needed promptly to meet the demand of real estate sales. They are also used to monitor radon test chambers in which other type of instruments are tested, inter-compared, evaluated and calibrated.

The disadvantages of continuous radon monitors are; 1) expensive compared to passive integrating devices with costs ranging from $1,000 to $8,000; 2) require a trained operator and 3) higher cost per measurement. About 5% - 10% of the radon measurements in the U.S. are made with continuous radon monitors.

There are four types of continuous radon monitors currently used by radon professionals.

1. Scintillation cell monitors: Figure 7
2. Pulse ionization chambers: Figure 7
3. Current ionization chambers: Figure 7
4. Solid State detectors: Figure 8

The devices must be calibrated initially at different conditions of exposure, recalibrated annually, and frequent cross-checks performed in the field to ensure accurate measurement results. The reliability of continuous radon monitors must be established and be maintained through a rigorous quality assurance program. They must meet the accreditation requirements of the NRSB or that of the NEHA-NRPP or a state equivalent program in order to be certified and be listed.

Figure 7. Continuous Radon Monitors

The sensitivities of different passive charcoal methods and continuous radon monitors are listed in Table 1. Test devices that have higher sensitivities can achieve results with smaller uncertainty.

Figure 8. Solid state detectors. Left-right: Durridge Rad-7, Radon Scout, Sun Nuclear 1028

Table 1. Different Measuring Instruments

Device				Detection Method	Net counts (CPM/ 4 pCi/L)
RTCA	2g	D-barrier	LS carbon	Alpha count	54.0
RTCA	50g	D-barrier	AC carbon	Gamma count	90.0
RTCA	90g	D-barrier	AC carbon	Gamma count	145.0
EPA	75g	D-barrier	AC carbon	Gamma count	48.0
PA	75g	D-barrier	AC carbon	Gamma count	60.0
RTCA	90g	Open-face	AC carbon	Gamma count	250.0
Sun Nuclear 1028		Passive		Solid State	0.17
Sun Nuclear 1029		Passive		Solid State	0.36
Radon Scout		Passive		Solid State	0.33
Durridge Rad 7		Active		Solid State	2.80
RS-300		Passive		Pulse Ionization	1.0
RS-500		Passive		Pulse Ionization	1.6
Femto 510		Passive		Pulse Ionization	1.2
Radalink Aircat		Passive		Pulse Ionization	1.7
Alpha Guard		Passive		Pulse Ionization	2.8
RTCA E-Smart		Passive		Current Ionization	1.2
Pylon AB-5		Active		Scintillation Cell	5.7

Table 2. Radiosensitivity of Electret Ion Chambers

Device			Volts/pCi/L/day
Rad Elec	0.2 liter	Short-term	2.0
Rad Elec	0.2 liter	Long-term	0.15
RTCA Radome	0.068 liter	Long-term	0.17

Table 3. Radiosensitivity of US Alpha Track Detectors

Device		LLD
Accustar	CR-39	0.3 pCi/L in 90 days
Landauer	CR-39	0.3 pCi/L in 90 days
RSSI	CR-39	0.1 pCi/L in 90 days

Table 4. Foreign Countries Alpha track Detectors

Country	Detection Material	Sensitivity (tracks/cm^2/4 pCi/L/day)
Germany	CR-39	3.6
Japan	CR-39	4.5
Ireland	CR-39	12.0
Italy	LR-115	22.0
NRPB (UK)	CR-39	9.2
Spain	CR-39	3.0
Sweden	CR-39	9.4

Conclusion

For short-term exposures for environmental radon (<4 pCi/L), diffusion barrier activated carbon collectors are the most sensitive and most cost effective devices that produce accurate results. The net counting rate of the charcoal devices is 25- 400 times higher than the net counting rate of the most sensitive to the least sensitive continuous radon monitors.

Electret Ion Chambers for short-term or long-term radon measurements require exceptional QA and QC steps to determine the contribution of background radiation when exposed at radon levels <2 pCi/L.

Alpha track detectors used for special measurements to determine the quarterly or annual average radon concentration can produce accurate results when using proper analytical procedures and the required QA and QC steps from the time the detector is produced until analysis.

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