

TEST METHODS FOR EVALUATING RADON BARRIERS

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ABSTRACT

It has been clear for some time that pressure-driven transport of soil gas through foundation openings is the major source for indoor radon. Diffusion of radon through these same openings and through the construction matrix is orders of magnitude smaller. While generally accepted practice for radon mitigation emphasizes sealing up radon entry routes, this is impossible in finished buildings. In new buildings, however, passive radon barriers are readily incorporated into the construction cycle, allowing us to design an impermeable barrier to block radon entry. Passive radon barriers may be considered in the form of membranes and films, as well as flowable material that is applied to the construction matrix. In practical terms, though, we cannot always expect to completely block the diffusive transport. Rather, the gas permeability of realistic radon barriers should be sufficiently low to block the pressure-driven transport while minimizing radon diffusion through the material. Additional factors necessary for defining optimum barriers include more general properties of durability, strength, and architectural compatibility. Three basic configurations are possible: (1) Test Specimens involving a benchtop apparatus and strictly controlled conditions in the laboratory, (2) Test Modules involving mock-ups constructed to mimic the final role of the material for laboratory or field trials, and (3) Test Buildings involving field trials of full-scale structures under controlled and naturally varying conditions. This paper reviews laboratory-based test methods that have evolved to measure the radon diffusion rate for candidate barrier materials.

INTRODUCTION

It has been clear for some time that advective transport of soil gas by pressure-driven airflows through foundation openings is the major source for indoor radon. Diffusive transport through these same openings and through the construction matrix is orders of magnitude smaller. Advective flows arise from depressurization effects of indoor-outdoor temperature differences as well as from secondary effects of operating heating, ventilating and air-conditioning (HVAC) equipment. Radon entry pathways are formed as the result of settlement and shrinkage cracks as well as from the design details of construction joints and service penetrations. For existing buildings, the preferred approaches to radon mitigation attack the pressure effects at the radon source, diverting radon entry to the open atmosphere: active subslab depressurization (ASD) systems (and variants to depressurize wall cavities) have reached a plateau of accepted practice. While ASD systems are generally effective, aesthetics, noise, energy consumption and necessary commitments to long-term maintenance stand in the way of complete acceptance.

While generally accepted practice for radon mitigation emphasizes sealing up radon entry routes, this is impossible in finished buildings. In new buildings, however, passive radon barriers are readily incorporated into the construction cycle. Ideally, an impermeable barrier is placed between the soil and the indoor airspace to block advective radon entry. Passive radon barriers may be considered in the form of membranes and films, as well as flowable material that is applied to the construction matrix.

We should not expect radon barriers to completely block the diffusive transport. We can, however, expect that they would block the pressure-driven transport while minimizing diffusive transport. We should also anticipate a secondary benefit from reduced water vapor transport. Radon barriers are not a primary control. Rather, they work in concert with the full building system. Such materials would be of particular value in bridging settlement and shrinkage cracks and other adventitious openings that form in the foundation and slab. In addition to commonly accepted materials currently in use as vapor barriers, alternative materials entering use in similar

applications warrant attention. A variety of geotextiles, organic polymers and bentonite clays, for example, have demonstrated practical efficiency as low permeability liners and caps in landfill applications (Grube, 1992).

The radon diffusion coefficient is the measurement variable of principal interest. Measurement strategies generally involve two chambers, one containing a radon source, the other separated from the radon source by the material undergoing the test. Three basic test configurations are possible:

- Test Specimen-- material samples are placed in a benchtop apparatus and tested under strictly controlled conditions in the laboratory
- Test Module-- mock-ups are constructed that integrate the test material into assemblies mimicking the final role of the material for laboratory or field trials under controlled and naturally varying conditions
- Test Building-- full-scale structures are constructed to integrate the material in final form for field trials under controlled and naturally varying conditions.

A family of ASTM Standard Test Methods has been developed for laboratory evaluation of diffusion resistance for protective clothing (e.g., F739), water vapor transmission through vapor barriers (e.g., E96, E398, F1249) as well as generalized gas permeability testing of plastic film and sheeting (ASTM 1434). These methods utilize small volume test cells that can be rack-mounted to allow for simultaneous testing of many specimens. This approach would be viable for simple specimens (e.g., plain sheet material), but would be difficult to adapt to articulated specimens (e.g., a caulked joint).

Diffusion cells are generally suited to testing small uncomplicated specimens. At some scale between the laboratory diffusion cell (size order of centimeters) and full-sized buildings (tens of meters), there is a role for benchtop modules capable of testing the net diffusion rate for barrier systems. While individual materials that become field-installed systems can be tested as separate specimens, the radon resistance for the system also depends on details of installation (e.g., formation of pinholes)--particularly at joints and seams. Methodology developed by Rogers and Associates (Kalkwarf et. al., 1982) and refined by Acurex (1994, 1995) allows for benchtop testing of pressure resistance as well as diffusion resistance for thicker material specimens such as concrete. Cylindrical core samples (4 inch diameter) are sealed into a special sleeve to constrain diffusion through the sample. Arnold (1990), on the other hand, developed a 1/30 scale model "house" (0.29 x 0.35 m) situated in a 1.07 m square box filled with sand. Detailed miniature taps were positioned to monitor the pressure fields associated with soil gas entry. Although Arnold's experiments did not specifically measure radon entry rates, extension of the design using source materials of sufficiently high radium content is straightforward.

For dynamic testing, McKelvey and Davis (1992) developed a materials testing chamber to evaluate radon permeation under controlled pressurization. The apparatus consists of a chamber (nominal volume, 340 L) with a constant radon source. The radon source features a calibrated pump to recirculate chamber air through the source material (uranium ore) and, if desired, to pressurize the chamber to a predetermined level. Housed within the chamber are cylindrical test cells (two sizes: nominal diameters of 10 and 15 cm) sealed at one end with the test material. Each test cell contains ports to allow for undisturbed radon sampling and pressure monitoring. The current design allows for up to three test specimens plus one control cell to be involved in any single test. Chamber testing can be conducted with the interior of the cells depressurized by as much as 50 Pa relative to the chamber. The chamber has been used to evaluate radon resistance for a complete range of barrier materials: caulks, cementitious materials, paints, foams, and membranes.

Larger test modules, in the form of rudimentary buildings, allow for focused evaluation of construction details under realistic conditions. Field-installed seams for sheet stock barriers, for example, could be evaluated on a temporary test pad using clean sand to simulate concrete loading. Test buildings, on the other hand, represent an ultimate test because construction details under test are usually integrated into a structure built for resale. Test protocols can be drawn directly from proven methodologies developed for new house evaluation studies (see, for

example, Williamson and Finkel 1994, GEOMET 1992). The most important step in field trials is locating the structure on a site likely to produce high levels of indoor radon. Consequently, protocols should embrace methodology to survey radon potential (see, for example, Tanner 1994, Yokel and Tanner 1992, or Rector 1991). Measurements of radon in the subslab, in the building (or test module) and outdoors are used to evaluate radon entry efficiency. Complementary measurements of tracer gases injected into the soil at the foundation establish soil gas entry.

MATERIALS AND METHODS

The theoretical and practical basis for laboratory testing of the diffusive resistance of membranes and films using small volume cells has been reviewed by Schwoppe et. al. (1988). The basic approach involves three main components (1) a source cell that is fed by the radon source, (2) the test specimen, and (3) a receiving cell to accumulate the radon that has diffused through the specimen. As shown in Figure 1, two design configurations are feasible: open loop and closed loop. In the open loop configuration, the receiving cell is constantly refreshed with radon-free air and the radon that has diffused through the test specimen is swept to the radon measurement system. Measurement systems for open loop configurations may be readily devised from continuous monitors (e.g., alpha scintillation, pulse-ion chamber). Alternatively, the radon swept from the receiving cell could be collected for deferred analysis (e.g., sorption onto activated carbon followed by gamma-ray spectrometry). In the closed loop configuration, radon diffusing through the test specimen accumulates in the receiving cell for measurement. The most simple measurements to support closed loop testing would rely on *in situ* detectors (e.g., passive electret, alpha-track detector). The source and receiving cells also could be integrated with electronic detectors, or sample air could be recirculated through an external detection system. While passive *in situ* detectors offer considerable cost reduction for closed loop testing, test results cannot be acquired without terminating (or at least interrupting) the test run. With external detection, test results are available throughout the test, but usually at the cost of more sophisticated instrumentation.

While a substantial body of published data and methodology exists for testing sheetstock materials for diffusive resistance to chemical agents, relatively few studies have been conducted for radon. In the early 1980s, Jha et. al. (1982) reported results for membrane testing with a closed loop apparatus consisting of a radon source (powdered uranium ore) separated from a scintillation flask radon detector by the membrane being tested. Radon monitoring continued until a constant count rate was evident, indicating steady-state conditions. The reported radon diffusion coefficients ranged from $10^{-9} \text{ cm}^2\text{s}^{-1}$ (0.7 mil mylar) to $10^{-6} \text{ cm}^2\text{s}^{-1}$ (2 mil natural rubber). Fleischer (1988) reported on a closed loop system composed of two small volume cups (a few hundred cm^3) separated by the membrane specimen. The lower cup held a radon emanation source (6-8 g uranium ore); each chamber contained an alpha-track detector to measure radon levels. The two cups were held together by a weight (approximately 1 kg); tests were 30 days in duration. Reported diffusion constants ranged from $10^{-10} \text{ cm}^2\text{s}^{-1}$ (polyethylene terephthalate) to $10^{-6} \text{ cm}^2\text{s}^{-1}$ (silicone-polycarbonate). The apparatus subsequently was applied to the problem of testing caulking materials (Fleischer 1992). Reported diffusion constants ranged from $10^{-9} \text{ cm}^2\text{s}^{-1}$ (Geocel) to $10^{-6} \text{ cm}^2\text{s}^{-1}$ (acrylic). Labed et. al. (1992) studied diffusion through extremely thin ($\sim 10 \mu\text{m}$) polypropylene membranes using a closed cell apparatus to deliver radon-bearing water to one side of the membrane while continuously monitoring radon levels in the air-filled volume over the opposing face of the membrane using an ion chamber detector. Although this test series was geared to development of a radon-in-water detection system, the underlying theme is compatible with general diffusion testing. One basis for open loop test for radon diffusion through various membranes was developed by researchers at the Lawrence Livermore Laboratory in the early 1970s (Hammon et. al., 1975). In the Livermore tests, a radium solution was used for the radon source, and radon that had diffused through the membrane into the receiving cell was periodically flushed from the cell with helium and swept to a molecular sieve trap maintained at liquid nitrogen temperatures. After sample collection was complete, the radon was desorbed at 350°C into a second liquid nitrogen-cooled trap and transferred to an alpha counter for analysis.

DISCUSSION

Laboratory determinations of the radon diffusion rate are predicated on radon measurements that represent transport by diffusion through the test material. The representativeness is reinforced by limiting the pressure gradient between the two cells, thus allowing diffusion to dominate transport. The one-dimensional diffusion equation is most often invoked to evaluate the results of laboratory testing in terms of the radon concentration (C , pCi L⁻¹), the diffusion constant for the barrier (D_e , cm² s⁻¹), the barrier thickness (L , cm), and the radon decay constant (λ , 2.1 x 10⁻⁶ s⁻¹):

$$\frac{\partial C}{\partial t} = D_e \frac{\partial^2 C}{\partial L^2} - \lambda C \quad (1)$$

Time-dependent and steady-state solutions that are specific to radon diffusion have been summarized by Collé et al. (1981), Rogers et al. (1984), and by Hart et al. (1986). General references on diffusion that provide additional guidance include the textbooks of Crank (1975) and Cussler (1984). For the laboratory test cell system, the radon concentration in the source cell (C_S , pCi L⁻¹) remains constant, and the concentration in the receiving cell (C_R) is a function of the cell volume (V , cm³), specimen area (A , cm²) and, for the open loop configuration, the airflow (Q , cm³ s⁻¹):

$$\frac{dC_R}{dt} = \frac{D_e A}{LV} (C_S - C_R) - \lambda C_R - \frac{Q}{V} C_R \quad (2)$$

Most laboratory tests strive to establish steady-state conditions so that the diffusion constant can be calculated from the ratio of concentrations receiving cell and the source cell.

$$\frac{C_R}{C_S} = \left[1 + \frac{(\lambda + \frac{Q}{V})LV}{D_e A} \right]^{-1} \quad \text{or} \quad D_e = \frac{C_R}{C_S} \left(\frac{(\lambda + \frac{Q}{V})LV}{A} \right) \left(1 - \frac{C_R}{C_S} \right)^{-1} \quad (3)$$

Components of the open loop configuration can be arranged so that radon that has diffused through the test specimen is accumulated to provide a measure of the radon flux (J , pCi s⁻¹). Under steady state conditions, the radon flux is related to the diffusion coefficient by:

$$J = C_S \frac{D_e A}{L} \quad (4)$$

The sampling apparatus integrates the flux over the collection period (t , seconds). Consequently, the accumulated radon (M_R , pCi) is the parameter of interest:

$$M_R = \int_0^t J dt = C_S \frac{D_e A}{L} t \quad \text{or} \quad D_e = \frac{L M_R}{C_S D_e t} \quad (5)$$

Regardless of the configuration (open versus closed loop; concentration versus flux) or the measurement technology selected for a particular test strategy, quality results are firmly tied to a thorough knowledge of the precision and accuracy of the measurement systems in use. This is not meant to imply that a certain level of precision or accuracy should be specified. Rather, the characteristics of the measurement system must be

integrated into the sampling design. Figure 2 summarizes steady state conditions in a receiving cell generated over a range of diffusion coefficients for a 1-mil membrane exposed to a source maintained at 1000 pCi L⁻¹. For a diffusion coefficient of 10⁻⁹ cm² s⁻¹, the concentration in the receiving cell would be 158 pCi L⁻¹. For a low flow open loop (one cell volume per hour, corresponding to a few mL min⁻¹), the steady state concentration is 1.4 pCi L⁻¹; if the open loop flow is fairly high (one cell volume per minute, corresponding to a few hundred mL min⁻¹), the radon concentration in the receiving cell is 0.02 pCi L⁻¹. A flux measurement, on the other hand, would collect 34 pCi in 24 hours. If the diffusion coefficient is lowered to 10⁻¹² cm² s⁻¹, results are lowered in all four systems. The closed loop would yield 0.2 pCi L⁻¹, concentrations in the open loop systems would be less than 10⁻³ pCi L⁻¹, and the flux system would collect only 0.03 pCi in 24 hours. Most commercially available radon measurement systems would perform adequately for the 10⁻⁹ cm² s⁻¹ case; for the 10⁻¹² cm² s⁻¹ case, however, only the closed loop configuration would seem feasible without greatly increasing the radon source level. Even then, extreme care would be required to discriminate concentrations that are in the same range as normal background.

It takes time to achieve the steady state conditions. For a 1-mil membrane characterized by a diffusion coefficient of 10⁻⁹ cm² s⁻¹, the relaxation time (the time needed to approach within a factor of e⁻¹ of the steady state) is about 10 minutes. The system would only require 30 minutes to move to within 95 percent of the steady state value. If the diffusion coefficient is lowered to 10⁻¹² cm² s⁻¹, however, the relaxation time increases to 76 hours. The system would require about 10 days to move to within 95 percent of the steady state value. A 10-day integrated concentration (if unadjusted for the ingrowth period) would under-estimate the steady-state condition by about 30 percent. Results from a 30-day integration, on the other hand, would be within a fraction of one percent of the steady-state value. Measurement strategies relying on integrating techniques (e.g., alpha track detector, passive electret), therefore must set a schedule that is long with respect to the relaxation time to reduce the impact of the ingrowth period on the final result. Shorter test periods can be considered if data is collected to support transient analysis. For integrating measurement systems, this could be accomplished with a sequence of integrated measurements.

Construction of radon sources is fairly straight forward. Source materials include uranium ores (see, for example Kalkwarf et.al. 1982), and radium salt solutions (see, for example Collé et. al. 1990). Even though radium is a naturally-occurring radionuclide, local jurisdictions require an approved health and safety plan supported by licensing and written procedures.

CONCLUSIONS

Radon barriers should become a useful adjunct to radon-resistant construction. Back-of-the-envelope calculations indicate that, taken alone, diffusion barriers of 10⁻⁹ cm²s⁻¹ or better would suppress indoor radon levels to a small fraction of the EPA-recommended action level of 4 pCi L⁻¹. Limited test data indicate that this could be readily achieved with existing materials. As interest grows in this area, however, formal tests should be considered to verify performance. Suitable technologies are already in place to support testing, and consensus standard methods should be developed.

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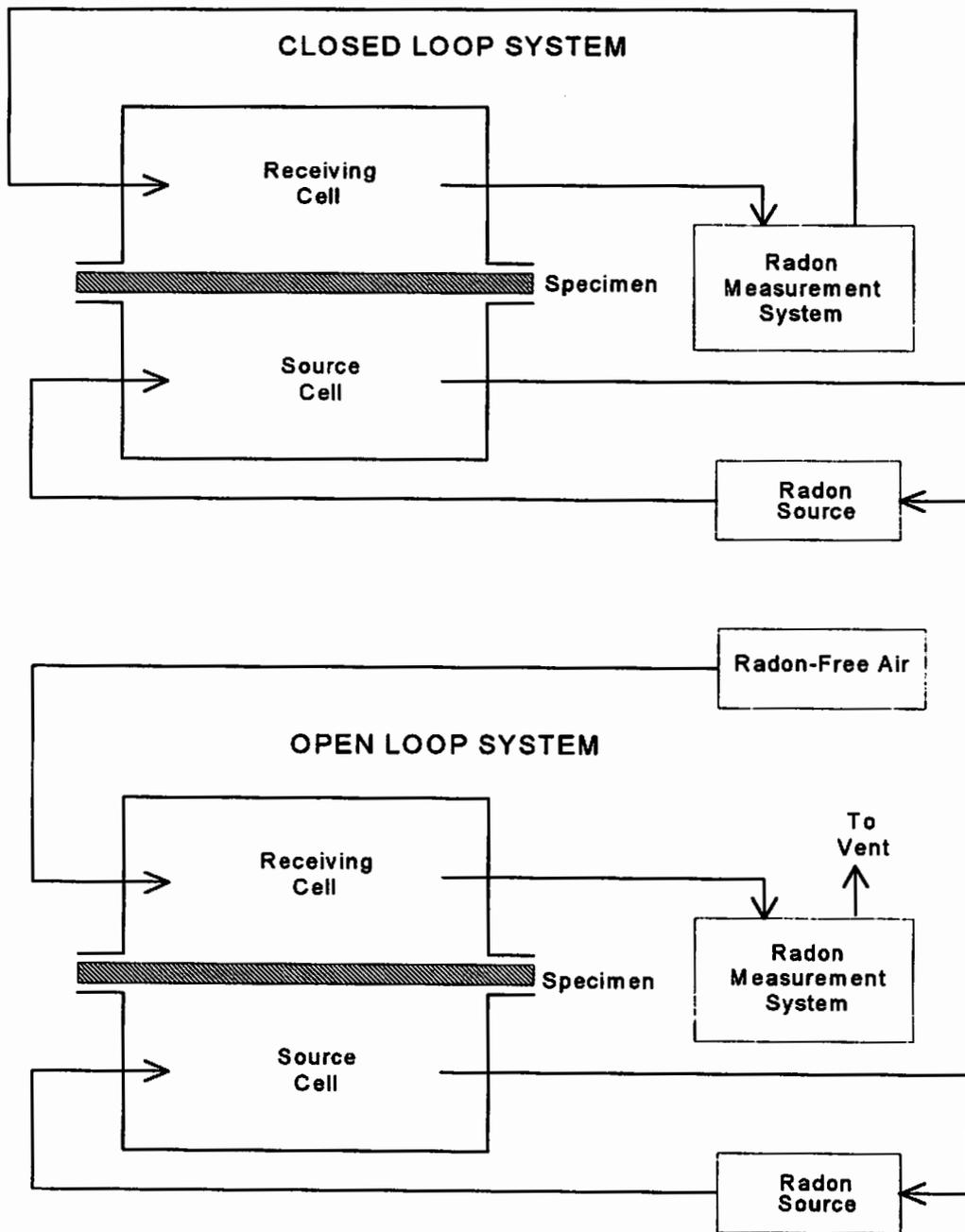
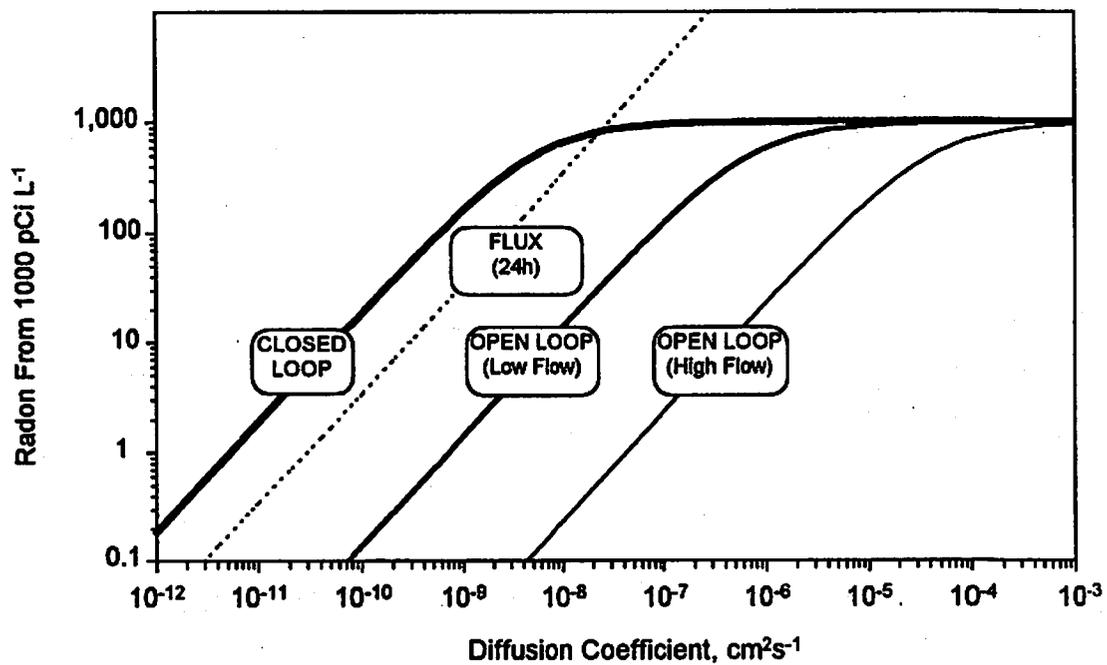


Figure 1. Closed Loop and Open Loop Configurations.



Plotted values represent conditions in the receiving cell given 1,000 pCi L⁻¹ in the source cell for a 1-mil membrane.

Figure 2. Variations Among Systems.