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MEASUREMENTS AND ANALYSIS OF THE TRANSPORT OF RADON THROUGH CONCRETE SAMPLES

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ABSTRACT

Experiments were conducted to measure the transportability of radon gas through common concrete samples which were characterized by their mix proportions, dimensions, porosity, air permeability and radon gas diffusion coefficient. Several innovative test systems and methods were designed, fabricated and calibrated to accurately measure these radon gas transport characteristics for concrete and to overcome many of the shortcomings of previously published experimental works. From the experimental results, it is found that diffusion is the dominant transport mechanism by which radon gas flows through an intact concrete slab. It is also shown that the indoor radon levels are greatly affected by the type of concrete mix employed. The results of this study can be utilized to improve the present technology of radon-resistant construction techniques for new residential construction.

INTRODUCTION

There are two major transport mechanisms that determine the amount of radon gas that enters a residential building: (1) the air pressure differential between the soil gas and the building air, more commonly known as advection and (2) the concentration gradient between the soil and the indoor air or mass diffusion. Both of these transport mechanisms are coupled to the natural properties of the intermediate media (normally concrete) which separates the two regions. The latter mechanism, diffusion of radon through the structure boundaries has generally been assumed to be negligible by the cement industry and more attention has been paid to radon ingression through the cracks and gaps in concrete slabs by pressure-driven air flow. Recent research on the transport phenomenon of radon diffusion has suggested that it is a significant contributor to indoor radon entry and that pressure-driven air flow is responsible for only 20 percent of the radon that enters a building from an underlying source (Kendrick and Langner 1991).

There are three main quantifying physical characteristics of concrete which are currently used to quantify concrete's ability to hinder the flow of radon. These include the porosity, the air permeability and the diffusion coefficient. The porosity of concrete is defined as the ratio of the void (air) volume in concrete to its bulk or overall geometric volume. An increase in the porosity will provide more air space within the concrete for radon travel, thus reducing its resistance to radon transport. The permeability of concrete classifies its ability to act as a barrier to radon gas movement when a pressure gradient exists across the concrete. The permeability is closely related to the porosity since an increase in porosity results in the size or quantity of void spaces available for air flow. The radon diffusion coefficient of concrete quantifies the ability of the radon gas to flow through concrete when a concentration gradient is the driving force. This parameter is also closely related and proportional to the porosity and permeability.

The most complete report on radon gas transport through concrete before 1981 is by Colle et al. (1981) while a more detailed summary of modern investigations is documented by Rosenberg (1993). Of the studies included in the aforementioned report and those later published in peer-reviewed literature measuring the characterizations of the radiological transport properties of concrete, the majority of works have reported large experimental uncertainties, ill-conceived experimental setups, invalid ranges for measurements or have demonstrated the lack of set standards in the field. To substantiate, a detailed summary of the most popular published works is now presented. The review is divided into two sections: diffusion of radon through concrete and air permeability coefficient (advective flow) of concrete.

Previous Work on the Diffusion of Radon Through Concrete

One of the earliest reported and most popular works on the flow of radon through concrete was by Culot et al. (1976) who experimentally measured the diffusion coefficient of radon through a basement-like structure that utilized uranium mine tailings as a backfill to create a radium source. A collection can was mounted against the wall to collect transported radon while pressure differentials across the wall and radon concentrations within the soil were monitored. Radon flux was determined by measuring the radon gas build-up within the can, but the instrument used to measure the radon concentration was not identified. There was also no reference to how steady-state conditions were obtained in the experimental procedure. Although the radon concentrations on the tailings side of the wall were monitored, nonuniformity throughout the tailings resulted in varying radon and moisture concentrations within the soil gas. Also, the soil-concrete interface was likely to experience varying radon concentrations due to the resistance of the soil to diffuse the soil gas. Finally, a linear diffusion model which forced its boundary conditions was used to determine an effective radon diffusion coefficient.

Another investigation into the diffusion of radon gas through concrete was performed by Zapalac (1983). An experimental apparatus was constructed to measure radon flux through concrete samples exposed to elevated radon levels. The radon gas which diffused through the concrete samples was accumulated in a collection loop and drawn from the collection can into an evacuated scintillation cell for measurement. Plastic bags were used in each loop in order to insure that a zero pressure gradient was held across the concrete sample while compressed air paint cans were used to accumulate the radon gas. Grab samples were taken from these cans to measure the radon gas present. The procedure used to create one-dimensional diffusion was not clearly explained, although it was stated that while the sealant around the concrete samples leaked, the design of the experimental apparatus limited the errors? Also, the paper stated that steady-state measurements were performed, but no explanation was made.

Further work concerning the problem of diffusion of radon gas through concrete was carried out by Folkerts et al. (1984). They measured the diffusion coefficients of radon through different building materials, including concrete. Their experimental apparatus was designed to count a portion of the alpha particles which were formed after the radon gas passed through the concrete. This measurement method was not independent of the elapsed time during the experiment because the progeny had previously plated-out on the sides of the collection chamber.

In Belgium, the exhalation and diffusion characteristics of three types of concrete mixes were experimentally determined by Poffijn et al. (1988). Concrete samples of the same mixture but of 1-10 cm thickness were found to have a great discrepancy in diffusion coefficients. It was found that the diffusion coefficients were three times greater for a 1 cm thick sample as compared to a 10 cm thick sample. Their discrepancies were probably due to poor fabrication of the concrete samples which lead to changes in their physical properties or to the experimental procedures which were not fully explained. The authors' final conclusion was that if their measured diffusion coefficients are used in determining the actual amount of radon entering a building by diffusion, an uncertainty of a few 100% should be used.

Rogers and Associates Engineering Corporation has done extensive research concerning the transport of radon through soil and concrete. One of their experimental studies (Rogers and Nielson 1991) indicated that a one-dimensional version of Fick's First Law was applicable to soil layers of different thickness. This assessment suggests that Fick's First Law may also be applicable in describing radon through concrete samples of differing thickness. In their latest work, Rogers and Nielson (1992) conducted experimental tests to determine the porosity, permeability, emanation and diffusion coefficients of nine different types of concrete typically used in building construction in Florida. Also reported was a correlation relating the water-cement ratio of concrete to its diffusion coefficient. This study has been found to be the most modern published work on radon diffusion through concrete.

Previous Work on the Air Permeability Coefficient of Concrete

One of the more influential studies on the radon advective flow in concrete was by Dimbylow (1987), who numerically solved the pressure-driven air flow equation for radon ingression through cracks in concrete floor slabs. The conclusion drawn from the study was that the low permeability of concrete will indicate pressure-driven flow through structurally intact concrete is unlikely to be an important pathway for radon ingression and that the sealing of cracks in concrete floors should be an effective remedial action to reduce the radon concentration in buildings.

More recently, McKelvey and Davis (1991) constructed a radon materials testing chamber to test the ability of various sealants to resist the advective flow of radon gas through the material under pressure. The system consisted of a pressurized chamber that contained radon gas to which the test sample was sealed. The radon gas which flowed through the concrete was collected and periodically measured. The results were expressed as an average reduction of the radon concentration within the pressurized chamber during an experimental run. Since four different sealants could be tested at the same time, comparative measures between the different types of sealants were reported. It appears that the system can easily conduct comparative tests, but can not quantify the actual radon flux through concrete due to a given pressure differential.

Additional work pertaining to the advective flow of radon through concrete was conducted by Kendrick and Langner (1991). This work like the investigations of Nazaroff et al. (1987) and Turk et al. (1991) estimated the radon flux ingression attributable to pressure-driven flow that entered through the cracks and concrete slab of a test structure. Kendrick and Langner's interpretation of the resulting data indicated that pressure-driven flow accounted for only a small fraction of the total radon entering the test structures through the test surfaces. Another study by Ruppersberger (1991) reported a simple procedure to select low air permeability concrete blocks. A general conclusion was that smooth-surface blocks usually are less permeable than blocks with a rough-looking surface. Finally, the previously mentioned work of Rogers and Nielson (1992) also presented simple empirical correlations for the air permeability coefficients of Florida concrete as a function of water-cement ratios. Their final conclusion was that advection through a concrete slab is negligible as compared to diffusion.

The purpose of this paper is to document the transport mechanisms that allow radon gas to pass through typical concrete building materials used in new residential construction. Experimental data is presented on the three main quantifying physical characteristics of concrete which are used to quantify the concrete's ability to hinder the flow of radon gas: the porosity, the air permeability and the diffusion coefficient. Concrete samples of different composition, including samples containing fly ash from Wisconsin coal fired power plants, were tested. Several innovative test systems and procedures which provide very accurate measurements are described. Finally, the present experimental results are compared with previously published data and are used to calculate the amount of radon gas which actually passes through an intact concrete slab by advective and diffusive forces.

EXPERIMENT

A description of the concrete test samples and of the test systems used in the experiments is now described. Complete details can be found in Rosenberg (1993).

Description of Concrete Samples

The size and shape chosen for the concrete samples were determined by the types of tests which were to be performed on the samples. A cylindrical shape was employed to simplify the process of pouring and sealing. The thickness that was selected allowed the radon diffusion tests to be performed at steady state over a reasonable period of time while still retaining a large enough thickness to represent the concrete's true properties. Test samples were fabricated according to Table 1 and several samples of each type of concrete mixture were fabricated. Sample A is a typical basement slab concrete mix (Hool 1918) while Sample B demonstrates how the substitution of fly ash changes the concrete's radiological transport properties. The last mix, Sample C demonstrates how an increase in water-cement ratio affects the concrete's ability to resist radon gas entry. The samples were prepared following the guidelines for laboratory concrete mixing preparation (U.S. DIBR 1966).

Table 1. Concrete mix proportions.

Sample	A	B	C
Parts Water	1	1	1.3
Parts Portland Cement	2	1	2
Parts Aggregate	3	3	3
Parts Sand	4	4	4
Parts Type F Fly Ash	0	1	0

The concrete samples were not cured according to the methods outlined by ASTM. These curing procedures were designed to obtain the maximum strength from a given concrete mix, while the present work desired to simulate the concrete which is present in a typical basement environment. Therefore, the concrete samples were kept moist on their surfaces for three days after casting and placed in a basement for the remainder of the curing period. This procedure closely simulated the curing conditions seen by an actual slab in a building. After the samples completely cured, their thickness and diameters were measured at 30 different positions with a micrometer. As a result of nonuniform shrinkage due to the different ingredients present in the concrete, the average thickness of the samples had a range of 2.692-3.056 cm.

The method of sealing the samples was determined by a trial and error process since previous work done in this area stated that problems occurred in sealing such that one-dimensional flow would occur. Our final method guaranteed that one-dimensional diffusion would develop since the gross radon flux would travel in the axial direction due to the sealing of the radial boundary. The concrete sample was originally cast in a split ring 25 mm thick with an inner diameter of 90 mm. After curing, the ring was removed and machined such that the inner diameter was 0.5 mm larger than the outer diameter of the concrete sample. To seal the sample in the holder, the inside of the steel ring was liberally coated with "Duro-Crete Steel and Concrete Epoxy." This epoxy demonstrated an ability to adhere to concrete and steel through visual inspection. It also provided a resistance to radon gas movement in tests that were later performed in the radon diffusion apparatus. The steel ring was then tightened around the sample until the mating portions of the ring came into contact with each other. After the epoxy had dried, the faces of the steel holder were machined parallel with an o-ring groove in each face. The sealed samples were then attached to the apparatus and visually tested for leaks. This procedure consisted of pressurizing one side of a sample which is immersed in water and visually inspecting for leaks at the steel-concrete interface. If leaks were detected, the steel ring was machined off and the sealing procedure was repeated until a leak-free seal was observed.

Description of Porosity Tests

Since the porosity of concrete is a good indicator of the transportability of radon gas, a method was developed to accurately measure this characteristic. Commercial porosimeters use mercury (a nonwetting fluid which can enter the concrete pores) at large pressures to determine the void (air) volume present in the concrete. This procedure makes the sample unusable for further testing. Another method consists of immersing the concrete sample in water and measuring its change in mass due to the water which has entered the pores. This method is inaccurate since the surface tension experienced by the water may not allow it to enter the smallest pores (ASTM 1992). Therefore, a procedure which did not have the shortcomings of the previously mentioned methods evolved.

A porosity system was designed utilizing Boyle-Marriott's Gas Law, which states that under isothermal conditions and low temperatures and pressures, a given quantity of a gas will obey the following relation:

$$\text{Pressure} \times \text{Volume} = \text{constant} \qquad (1)$$

Figure 1 illustrates the porosimeter used in our experiments. It consisted of two air chambers which were monitored with temperature and pressure sensors hooked-up to a PC-data acquisition system (PC-DAS). The larger chamber with a volume of V_1 contained the concrete sample (before it was sealed in its steel ring). The exhaust chamber with a volume of V_2 was used to measure the amount of air present in the concrete's void spaces by exhausting the pressurized source chamber into the smaller chamber.

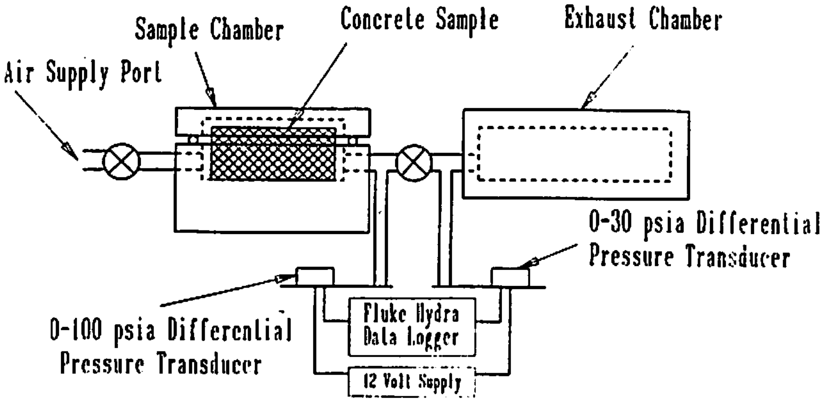

Fig. 1. Schematic of porosity measurement system.

After the concrete sample was placed into the source chamber, the chamber was sealed and pressurized to 100 psi. The pressure within this chamber was then observed to decrease until the pores within the concrete were at a pressure equal to the air in the chamber. At this point, the pressure in the exhaust chamber (P_0) which was near atmospheric pressure was recorded. The larger chamber was then exhausted into the smaller one and the final equilibrium pressure (P_2) within the two chambers was recorded. From Boyle-Marriott's Gas Law, an equation was formulated which creates a mass balance of the air within the chamber. By simple algebraic manipulation, the solid volume of concrete (V_S) can be solved by:

$$V_S = [P_2 (V_1 + V_2) - P_0 V_2 - P_1 V_1] / (P_1 - P_2) \tag{2}$$

By knowing the geometric or bulk volume of the concrete (V_B) from the thickness and diameter measurements, the porosity can then be calculated as:

$$\varepsilon = (V_B - V_S) / V_B \tag{3}$$

To insure that the volume of the sample chamber was the same for each experimental run, a shim was placed under the sample volume cover to guarantee that it was tightened down to the same position each time. In order to calibrate the system, a steel slug of known diameter and zero porosity was inserted into the chamber and the experimental procedure was repeated. Ball bearings of 0.25" and 0.5" diameters were then added to the steel slug in the chamber to provide a simulation of the solid volume of each concrete sample (V_S). By repeating the procedure with the concrete samples and comparing the results to those received from the calibration runs, the solid volume of each sample could be determined. An overall uncertainty of 1.5% was calculated for these porosity measurements. The details of the uncertainty analysis are described in Rosenberg (1993) and are not repeated here for brevity.

Description of Air Permeability Tests

Since the advective flow of radon gas through concrete has been recognized as an important transport mechanism by which radon enters a house, an air permeability test was designed which would measure the resistance of concrete to a pressure-driven air flow. Current permeability tests only measure the permeability of concrete to water or they describe a relative permeability measurement due to the ability of the concrete to resist an electrical charge. Other permeability tests are carried out at higher pressures than those associated with the actual pressure differential (1-10 Pa) seen by concrete in a building (ASTM 1992). These results are then used to estimate the air flow at lower pressure differentials. No standard test procedure has been documented to exclusively measure the permeability of concrete with respect to air flow.

The present permeability apparatus which could measure the volumetric flow rate through the concrete samples subject to a pressure differentials greater than 1000 Pa is illustrated in Fig. 2. The simple design of the system was based upon the following assumptions: (i) radon and air molecules coexist with zero slip velocity and (ii) since radon is an inert gas there would be no chemical reaction within the concrete sample that would hinder the flow of radon. The air permeability coefficient (K) is defined by a modified Darcy's Law (Dullien 1979) as:

$$V = (-K\, P_{avg} / \mu\, P_2)\,(dP/dx) \tag{4}$$

where,

V = volumetric air flow rate
K = permeability coefficient
P_{avg} = average air pressure in the concrete sample
P_2 = air pressure at the face of the concrete sample where air is exiting
μ = dynamic viscosity of air
dP/dx = pressure gradient across the sample

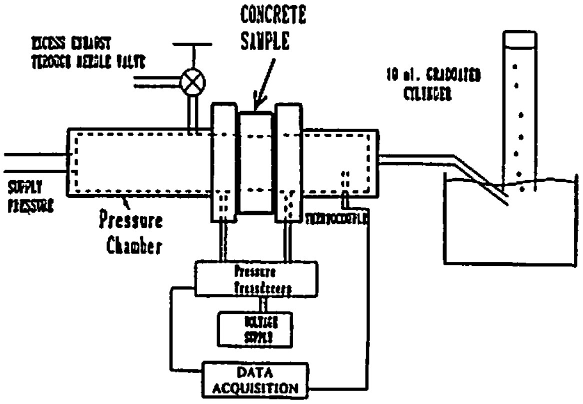

Fig. 2. Schematic of high pressure air permeability measurement system.

To create a pressure differential across the concrete sample, two valves were placed at the inlet and outlet of the pressure chamber. With these valves the air flow through the chamber could be kept very small to minimize the bulk fluid motion of air within the pressurized chamber. Two differential pressure sensors were then used to measure the pressure at each face of the concrete sample and a thermocouple was utilized in the accumulation chamber to monitor the air temperature, hence the dynamic viscosity. Both of these sensors were connected to a PC-DAS that employed the commercial data acquisition software "Labtech Notebook." To measure the air flow rate through the concrete, a graduated cylinder was filled with distilled water and inverted in a water reservoir. The volumetric flow rate was then determined by monitoring the amount of water displaced by air in the graduated cylinder over a given period of time.

To measure the air permeability of the concrete samples at pressure differentials below 1000 Pa (at lower pressure differentials the surface tension created at the air-water interface in the graduated cylinder could not be overcome to form a bubble), a modified system (Fig. 3) was employed. The only significant change in the experimental setup was the sealing of the accumulation chamber, which allowed the air to pass through the concrete to be collected for measurement. By utilizing the perfect gas law and by monitoring the changing pressure (ΔP) within the accumulation chamber, the mass accumulation rate within the chamber (Δm/time), which is the mass flow rate of air through the concrete can be calculated by:

$$\Delta m/\text{time} = (V/RT) \times (\Delta P/\text{time}) \tag{5}$$

where,

m = mass of gas in the accumulation chamber
P = pressure in the accumulation chamber
R = specific gas constant of air
T = temperature in the accumulation chamber
V = volume of the accumulation chamber

To substantiate that a steady-state pressure gradient existed across the concrete sample, experiments were carried out to determine the length of time before steady conditions were prevalent after the chamber was pressurized. By using the standard propagation of error method, an experimental uncertainty in the air permeability coefficient of +/- 8% was calculated.

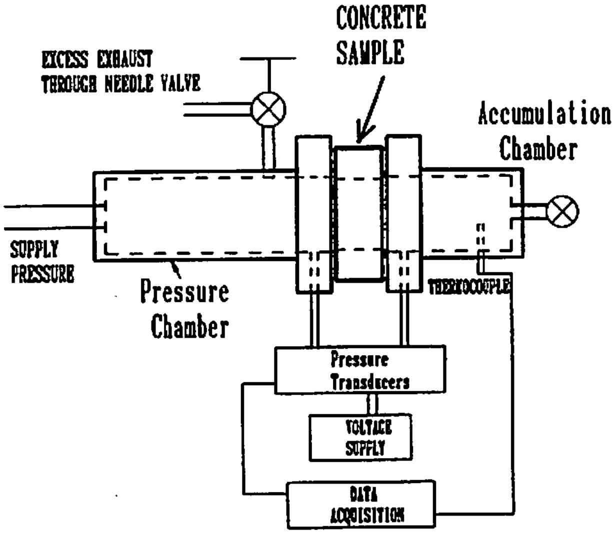

Fig. 3. Schematic of low pressure air permeability measurement system.

Description of Radon Diffusion Tests

The ability of the concrete to act as a barrier to radon diffusion is quantified by its diffusion coefficient which is defined by Fick's Law. Fick's Law as applied to a slab of concrete experiencing one-dimensional fixed concentration differences with isobaric and isothermal conditions is expressed as:

$$J = D_{RC}\, dC/dx \tag{6}$$

where,

D_{RC} = diffusion coefficient of radon through the concrete
J = radon flux through concrete per cross-sectional area
dC = concentration difference across the concrete sample
dx = thickness of concrete sample

In order to determine the diffusion coefficient of radon through a given concrete, the radon which passes through the concrete over a period time due to the concentration difference must be measured. The apparatus that accomplished this is schematically drawn in Fig. 4.

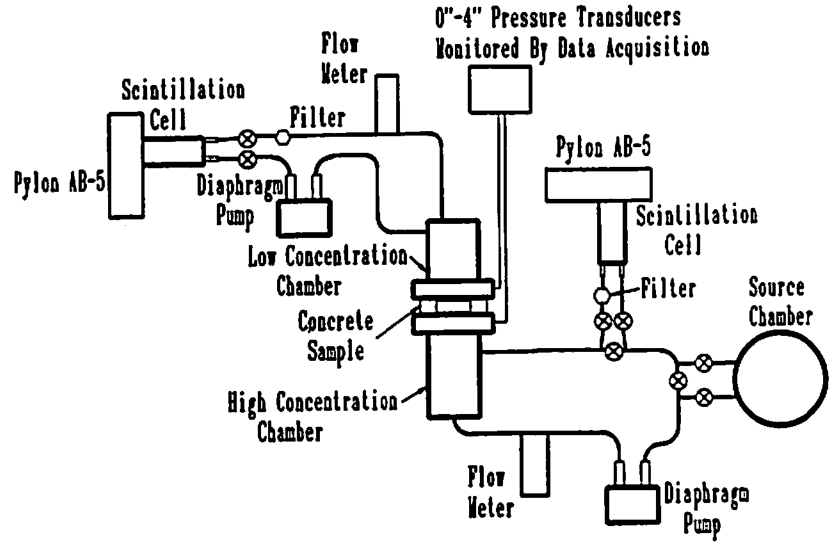

Fig. 4. Schematic of radon diffusion measurement system.

Two calibrated Pylon Model AB-5 Portable Radiation Monitors with Pylon Model 300A Lucas Cells were used to measure the radon concentrations within the diffusion system. The Lucas cell allows radon laden air to continuously pass through it while the Pylon AB-5 registers the counts received from scintillating particles. The components in each loop are connected by 0.25" ID Tygon tubing. A diaphragm pump is used in each loop to assure that the air and radon in each loop is thoroughly mixed. A 0-5 liter per minute flow meter is used in each loop to monitor the flow rate since the calibrated sensitivity of the radon monitor is dependent on the flow rate. An accumulation chamber is attached to each face of the concrete to allow the full facial area of each concrete sample to be exposed to the radon-air mix. A filter is placed at the entrance of the scintillation cells to remove dust and radon daughter products within the air stream. Toggle valves and various hardware are also employed to create the desired flow configuration while a PC-DAS is connected to the pressure transducers and the Pylon AB-5 for monitoring and data collection.

The diffusion system was pressurized and tested for leaks. This was accomplished by placing a small amount of Freon-113 in the source chamber and circulating it throughout both loops. The system was then pressurized to 5 psi with an external air supply and a T.I.F. 5500 Pump Style Automatic Halogen Leak Detector was used to locate leaks.

In order to accurately measure the radon flux and concentration gradient which occurred during the diffusion process, an innovative procedure was developed. This procedure did not rely on continuous monitor measurements, but instead utilized an instantaneous grab sampling technique to measure the radon gas concentrations present. It was discovered through preliminary experiments that the continuous monitoring technique creates a large discrepancy between the actual activity present and the measured activity because of the radon daughter plate-out within the Lucas cell. This problem had been recognized by previous users of scintillation cells, most notably by Countess and Thomas (1977) and Busigin et al. (1979) who tried to mathematically correct the counts received from the continuous scintillation cell monitor. The grab sampling technique is not affected by the daughter plate-out phenomena if the calibration procedure for radon grab sampling is similar to the steps followed to obtain an experimental measurement within the experimental apparatus. The scintillation cell monitor combination is calibrated by taking in a known concentration of radon gas and allowing it to reach an equilibrium level with its daughter products. After 3.5 hours (the time needed to reach equilibrium) the scintillations counted by the monitor can be converted to a radon concentration factor by calculating the efficiency of the cell as follows:

$$E = (NCPM \times F_1) / (2.22 \times 3 \times A \times F_2) \qquad (7)$$

where,

NCPM = Net Counts Per Minute registered by the monitor
F_1, F_2 = correction factors for decay during the counting interval
A = activity in the scintillation cell in pCi

The 2.22 relates decays per minute to pCi and the 3 is the number of alpha emitters in the radon decay chain. It can easily be seen that if the same counting intervals are used for calibrating and actual measurements, a conversion factor can be formulated which converts the counts per minute directly to the radon activity present. This method was used to calibrate the monitors with the calculated efficiency remaining constant at varying radon gas concentrations.

The experimental procedure for diffusion began by allowing radon gas to accumulate in the source chamber for a given period of time. The gas is then circulated through the loop containing the source chamber, therefore creating a concentration gradient across the sample. The pump is then turned off and the radon was allowed to diffuse through the concrete sample for a measured period of time, after which a grab sample measurement is taken from each loop. From this experimental procedure, the radon present in each loop is known at the beginning of each experiment by calculating how much radon had been emitted by the radium source. The amount of radon present in the loops due to the diffusion of the radon through the concrete is determined from the grab sample at the end of the experiment. By repeating this experiment with different time periods for diffusion, the diffusion coefficient of radon through the concrete sample was determined.

To determine steady-state conditions across the concrete sample, a set of experiments were run at the same initial concentration level but with incremental periods of diffusion. Experiments were run with a diffusion time of one hour and then repeated with the diffusion periods increasing by one hour increments. When the diffusion coefficient which was calculated for each subsequent time interval did not increase from one hour to the next, steady state was defined and the diffusion coefficient of the tested sample was recorded.

Another problem which may occur due to the air not being mixed is a settling of the radon gas to the bottom of the chambers. In order to determine the magnitude of this effect, a set of diffusion experiments were conducted. Each experiment was exactly repeated except for the orientation of the concrete sample with respect to the earth's gravitational field. It was assumed that if there was settling of the radon gas within the chambers due to gravity, the orientation of the sample where gravity works in the same direction as diffusion would result in a larger diffusion coefficient than the orientation where gravity works against the diffusion process. The diffusion coefficient calculated with gravity neither assisting or hindering the diffusion process would then lie between these two values. The results of these experiments indicted that settling did not occur within the chambers and that a uniform concentration gradient existed at the face of the sample.

Although the radon settling tests indicated that a uniform concentration existed across the faces of the concrete sample, it did not ensure that a uniform concentration existed throughout the accumulation and collection chambers. The radon concentration measurements which were taken from each chamber yielded the average level of radon activity in the chamber, not the actual concentration level present at the face of the concrete sample. While slight concentration gradients in the chambers will exist due to the diffusion occurring through the concrete sample, it is assumed that the diffusion coefficient of radon (on the order of 10^{-1} cm^2/s) is large enough to ensure a uniform concentration will exist. An experimental uncertainty of +/- 10% in the diffusion coefficient measurements was calculated (Rosenberg 1993).

DISCUSSION OF RESULTS

Experimental Results

A summary of the experimental results obtained in this investigation are displayed in Table 2 which documents the porosity, the air permeability coefficient and the diffusion coefficient for two samples of three different concrete mixtures.

Sample A, which was chosen to simulate a typical basement slab concrete mix has a porosity similar to the values previously reported for a "normal" concrete mix. The addition of fly ash in Sample B resulted in an unexpected increase in the porosity. A number of previous investigations have reported substantial decreases in the porosity value when fly ash was substituted as an ingredient. Further study of this discrepancy determined that by removing 50% of the cement and replacing it with fly ash results in insufficient hydration (solidification) during the curing process. The Type F fly ash which was used in these experiments does not solidify without the assistance of a proper amount of Portland Cement. Typically, fly ash varies greatly in its chemical composition due to the combustion products which created it, thus proper concrete mix specifications must be followed (ASTM 1992). The expected increase in porosity of Sample C occurred since the excess water which was available during the curing process created water filled voids. As the concrete dried, the excess water migrated to the surface of the concrete where it evaporated. The migration creates tortuous routes through the concrete which results in the increased porosity and allows easy passage for radon gas through the concrete.

The porosities of the tested concrete samples ranged from 0.12 to 0.20 when comparing Sample A to Sample C. This approximate 70% increase translates into a greater volume of void space (air volume) in the concrete through which radon can freely travel. It is not known if this increase in porosity is due to an increase in the average pore size, an increase in the amount of pores, or a combination of both of these factors. What has been shown is that the amount of air space for radon gas transport can be controlled significantly by the selection of concrete mixture proportions.

The air permeability coefficients which are presented in Table 2 are an average value calculated from the low (<1000 Pa) and high (>1000 Pa) pressure tests. The low pressure experiments displayed a higher permeability coefficient for experimental runs of increasing length since the decreasing pressure differential did not cause a direct corresponding decrease in the flow rate through the concrete sample. This was due to the time required for an equilibrium level to exist across the concrete sample after it had been exposed to a given pressure differential. The average values of all the tests are close to the average value which was measured from the high pressure tests alone.

The individual values of permeability follow the same trend as the porosity measurements. The air permeability coefficients for Sample A are the lowest while Sample B values are the largest (a 270% difference). The air permeability coefficient for Sample C is approximately 123% greater than those exhibited for Sample A.

As expected the measured diffusion coefficients as documented in Table 2 correlate well with the trends seen in the porosity and permeability experimental values. Again the Sample A concrete (normal concrete mix) yielded the lowest values of diffusion coefficient while Samples B and C had values that were approximately 189% and 83% greater, respectively.

Table 2. Summary of Experimental Results.

Porosity	Permeability [cm^2]	Diffusion Coefficient [cm^2/s]	Comments (Reference)
0.12	1.45×10^{-12}	4.38×10^{-4}	A1 **Present Work**
0.13	1.24×10^{-12}	5.53×10^{-4}	A2
0.2	5.42×10^{-12}	14.7×10^{-4}	B1
0.19	4.52×10^{-12}	13.9×10^{-4}	B2
0.17	3.03×10^{-12}	8.86×10^{-4}	C1
0.17	2.98×10^{-12}	9.32×10^{-4}	C2
0.06	X	1.69×10^{-5}	Calculated Porosity (Culot et al. 1976).
0.1	X	6.8×10^{-6}	Tested "Heavy" Concrete (Folkerts et al. 1981).
0.07	X	3.34×10^{-4}	Concluded that radon entry through slab is negligible (Siotis and Wrixon 1984).
0.32	X	6.01×10^{-4}	
0.11	X	2.83×10^{-4}	Stated that an "uncertainty of a 100% existed in these values (Stranden 1983).
0.13	X	4.7×10^{-4}	
0.11	X	1.69×10^{-4}	
0.2	X	1.0×10^{-3}	Diffusion coefficients were measured using transient method (Countess and Thomas 1977) which was formulated for a steady-state diffusion coefficient (Rogers and Nielson 1992).
0.21	X	2.9×10^{-3}	
0.26	4.4×10^{-12}	1.3×10^{-3}	
0.26	3.4×10^{-12}	1.2×10^{-3}	
0.22	3.6×10^{-12}	4.6×10^{-3}	
0.26	4.0×10^{-12}	3.3×10^{-3}	
0.23	X	3.9×10^{-3}	
0.17	6.5×10^{-12}	6.3×10^{-4}	
0.18	8.0×10^{-12}	2.2×10^{-4}	
0.17	8.7×10^{-12}	1.8×10^{-4}	

Table 2 also contains a comparison of the present data with other published results. While it is difficult to directly compare these results due to the many different types of concrete that were tested, the porosity measurements do indicate concrete mixes with similar radiological transport properties. A comparison of air permeability and diffusion coefficients indicates relative agreement (same order of magnitude) between the present findings and those listed in Table 2 for like porosity values. In the present work, the permeability and diffusion coefficients follow the same trend as the porosity (an increase in porosity generates an increase in the permeability and diffusion coefficients), while the findings of Rogers and Nielson (1992) are somewhat inversed in that increases in porosity do not necessarily produce higher permeability and diffusion coefficients. This trend is probably the result of employing transient measurements to calculate steady-state coefficients.

In order to determine the applicability of Fick's Law in modelling the diffusion of radon through the concrete samples at varying concentration levels, another set of experiments were run at different concentration levels. Table 3 which displays the results for a Sample A mixture shows that even at varying concentration levels, Fick's Law applies to the diffusion of radon through concrete.

Table 3. Experimentally measured diffusion coefficients at different concentration gradients.

Concentration Gradient [pCi/L]	Diffusion Coefficient [cm^2/s]
92	4.3×10^{-4}
463	4.5×10^{-4}
554	4.0×10^{-4}
583	3.9×10^{-4}
596	4.0×10^{-4}
605	4.0×10^{-4}
664	4.3×10^{-4}
1,129	4.2×10^{-4}
2,300	4.0×10^{-4}

Several previously published works have concluded that Fick's Law doe not hold for concrete samples of varying thickness. A second set of concrete samples with the same mixture ingredients but a 5 cm thickness were poured. The results as shown in Table 4 for a diffusion test indicate that the thickness of the concrete sample does not affect the diffusion coefficient.

Table 4. Diffusion coefficients for concrete samples of different thickness.

Sample Thickness [cm]	Diffusion Coefficient [cm^2/s]
3	1.2×10^{-3}
5	1.3×10^{-3}

Application of Experimental Results

An example is now provided to show the magnitude of the radon gas entry rate for an intact concrete slab of a residential basement due to the two dominant transport mechanisms (advection and diffusion).

For the advection calculation (Fig. 5), we utilize Darcy's Law and the following values:

$dP = 4$ Pa
$dx = 10$ cm (typical basement slab)
$K = 1.45 \times 10^{-12}$ cm^2 (experimentally determined for Sample A1)
$\mu = 1.846 \times 10^{-5}$ N s/m^2 (at room temperature)

where dP is a typical pressure differential across a basement wall during the winter heating season (Renken and Konopacki 1993). The result is a volumetric flow rate of 3.14×10^{-7} L/s/m^2 that will pass through the concrete floor. In a typical basement with a 140 m^2 (1500 ft^2) slab this would produce an overall flow of 0.157 L/hr of air passing through this concrete slab. With the soil gas typically containing 1288 pCi/L (Carlisle and Azzouz 1991), this will result in a radon entry rate of approximately 203 pCi/hr. If this radon gas is diluted in a house with an overall volume of 1000 m^3, this radon entry rate will produce 2.04×10^{-4} pCi/hr which will be received by each liter of air in the house. Finally, if the decay of radon is the only mechanism which is removing the radon laden air, the equilibrium level of radon due to the advective entry alone is 0.03 pCi/L.

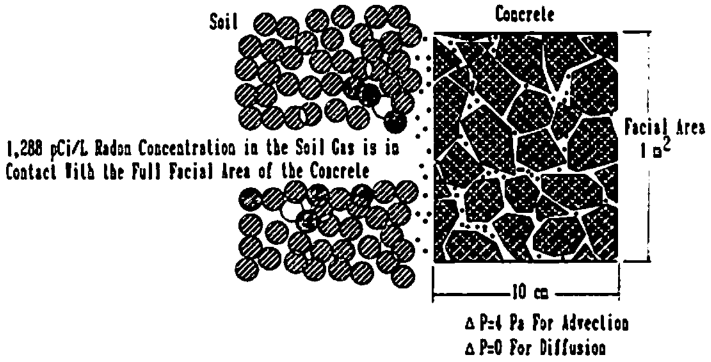

Fig. 5. Conditions present for advective and diffusive flux calculations.

The conditions of the diffusion calculation are also given in Fig. 5. By using Fick's Law eqn (6), experimental values of the diffusion coefficient, and the following values:

$dC = 1288$ pCi/L (same soil gas activity as advective entry example)
$dx = 10$ cm (typical basement slab)

a radon entry flux of 2030 pCi/m^2/hr is calculated to pass through the concrete slab. In the same fashion as the advective entry rate calculation, 0.28 pCi/hr will be received by each liter of indoor air. If the radon decay is the only mechanism which removes the radon from the indoor air, the equilibrium level of radon will be 37.2 pCi/L. In order to more accurately determine the radon activity level in a building, the ventilation rate would be included as a radon removal mechanism. It is extremely difficult to determine the affects of the ventilation rates due to three important factors: (1) air exchange rates in homes vary widely from 0.1 to 1 per hour, (2) the air entering can be radon free outdoor air or radon laden soil gas, and (3) the actual air movement in a house varies from room to room and between levels. If the ventilation rate of 0.5 hr^{-1} is chosen, an equilibrium level of 18.6 pCi/L will exist in the house. This indicates that the diffusion of radon gas through concrete is a significant source of radon in indoor air.

It also illustrates that under the prescribed conditions, the diffusive contribution to elevated indoor levels is many times greater (in this case 620 times more) than the advective contribution.

The above application assumed the same radon activity levels on the soil side of the concrete slab. This may not occur in an actual building where the pressure differential which exists across the slab may extend into the soil, pushing more of the radon which is generated within the soil towards the wall. A recent theoretical study (Rogers and Nielson 1991) which takes this into account by using experimentally measured permeability and diffusion coefficients to solve the transport equation, included this effect of the pressure gradient extension into the soil. The results indicated that in soils with low permeabilities the diffusive radon flux was 100 times the advective radon flux. If a highly permeable medium such as sand is used as the base for the concrete slab, the diffusive flux was found to be twice the advective flux. These results ratify our findings that the diffusive flux through an intact concrete slab in an actual building is much greater than the advective flux.

CONCLUSIONS

This paper discussed a series of experiments that were conducted to measure and quantify the transport mechanisms which allow radon gas to penetrate through concrete building materials and cause an unhealthy indoor radon level. Values of porosity, air permeability and diffusion coefficients were presented for three different concrete mixes. The experimental systems and procedures documented in this study were developed to overcome the shortcomings of previous works and to minimize the experimental uncertainty. The experimental findings showed: (1) the radiological transport characteristics with regards to concrete acting as a radon barrier can be controlled by the type of mix proportions and ingredients and (2) diffusion of radon gas is the dominant transport mechanism in an intact concrete slab.

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