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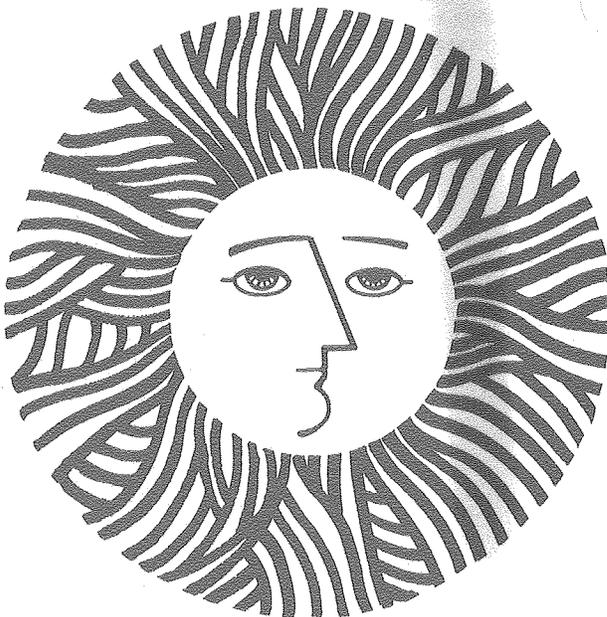
A TIME-DEPENDENT METHOD FOR CHARACTERIZING THE
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Geordie H. Zapalac

July 1981

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A TIME-DEPENDENT METHOD FOR CHARACTERIZING THE DIFFUSION
OF RADON-222 IN CONCRETE

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ABSTRACT

The porosity and diffusion length of concrete have been determined by measuring the time-dependent diffusion of radon through a thin slab of the material. One surface of the slab is exposed to a large, fixed radon concentration beginning at $t=0$. The radon that diffuses out of a portion of the opposite surface is collected during several contiguous time intervals. The total activity collected over a set of intervals beginning at $t=0$ and the steady-state flux of activity are used to calculate the porosity and diffusion length. As a test of these parameters, they are then used to predict the activity collected during other time intervals and for other sample thicknesses. Samples from two types of concrete were tested: one type yielded a porosity of 0.068 and a diffusion length of 12.6 cm; the respective values for the other were 0.32 and 16.9 cm. The predicted and experimental results agreed well, thereby verifying the assumption that concrete may be treated as a homogeneous diffusion medium for radon.

Keywords: concrete, diffusion length, porosity,

radon.

INTRODUCTION

Radon concentrations in soil gas have been commonly observed in the range of 300-2000 pCi/l (picocuries/liter) (Is62). Diffusion of radon through the concrete slab under a house may constitute a significant source of occupant exposure to radon and its decay products.

Concrete is a complex, heterogenous material. The assumption is often made, however, that concrete may be treated as a homogeneous diffusion medium for radon (Cu75;Jo78;Kr71). Based on this assumption, its diffusion is characterized by two parameters: the diffusion length ℓ and the porosity ϵ . The diffusion length is approximately the distance required for a steady-state gradient in a sourceless medium to decay to a factor of $1/e$. The porosity is the fraction of volume of the concrete occupied by air.

Previous determinations of diffusion parameters have often required time-consuming measurements, and difficulties have been encountered in attempting to determine ℓ and ϵ independently. Jonassen and McLaughlin placed a large concrete slab in a pressurized vessel and measured the radon generated by trace amounts of radium-226 occurring naturally within the sample (Jo78). After several weeks they measured the equilibrium concentration of radon in the chamber. Because ℓ varies with pressure and ϵ does not, the authors were able to separate ϵ and ℓ by repeating these measurements at different pressures. The values they found were $\ell=7.44$ cm and $\epsilon=0.265$. Culot, et al constructed a room of concrete and banked uranium mill tailings against the outside walls (Cu75). The concentration of radon on both sides of the walls provided boundary conditions for the steady-state solution of the diffusion

equation. The flux of radon into a shallow container sealed upon the inside wall was measured. This measurement does not separate ϵ from ℓ . Assumptions of $\epsilon=0.25$ and $\epsilon=0.05$ would fix Culot's value of ℓ at 7.6 and 12.7 cm, respectively. Krsiuk et al sealed a sample on all sides but one and covered the exposed surface with a chamber (Kr71). The increase in activity within the chamber was measured by sampling the radon concentration every 24 hours over a period of two weeks. The ratio of interstitial volume in the sample to the volume of the chamber was assumed to be 0.1, although the volumes of the chamber and sample were not reported. This assumption yields diffusion lengths ranging from 13 cm for heavyweight concretes to 29 cm for lightweight concretes.

In this paper a rapid, laboratory-scale alternative to these methods is described. The time-dependent diffusion of radon through a thin disc of concrete cut from a standard test cylinder is observed. One surface of the sample is exposed to a fixed concentration of radon beginning at $t=0$. The radon emerging from an area on the opposite surface is collected and measured as a function of time. The rate at which the flux approaches its equilibrium value depends upon ℓ alone, although the gross flux depends upon both ℓ and ϵ . A measurement of the total activity collected during a time interval before the system has reached steady-state and a measurement of the steady-state flux are sufficient to determine ℓ and ϵ . These parameters were determined for two types of concrete and successfully used to predict the radon collected for different time intervals and thicknesses, confirming the previously unvalidated assumption that concrete may be treated as a homogeneous diffusion medium for radon. Measurement of ϵ and ℓ can be completed in one day. At least two measurements on each sample assure their reproducibility.

THEORY

The diffusion of a substance through a homogeneous medium is described by Fick's law and the diffusion equation:

$$\vec{j} = -D\nabla C \quad (1)$$

$$D\nabla^2 C - \lambda C = \frac{\partial C}{\partial t} \quad (2)$$

Although equations (1) and (2) are not independent, it is convenient to consider them separately. In the equations:

C is the activity of radon per unit volume of interstitial air space in the concrete,

j is the flux of activity per unit area across a plane within the interstitial volume,

λ is the decay constant for radon ($7.58 \times 10^{-3} \text{ h}^{-1}$), and

D is the effective diffusion coefficient of radon within the material. The diffusion length is defined from the diffusion coefficient as $D = \lambda \ell^2$,

Other investigators often define the diffusion coefficient as D/ϵ (Jo78, Kr71).

No source term appears in (2). The generation of radon within the concrete from trace amounts of radium is on the order of 0.1-2.0 pCi kg⁻¹ hr⁻¹ (In81a). This amount is negligible in this experiment.

In the case of a slab, the diffusion profile is one-dimensional away from the edges and (2) reduces to:

$$\partial^2 C / \partial x^2 - C / \ell^2 = \frac{1}{D} \partial C / \partial t \quad (3)$$

Consider a concrete slab of thickness L underlying a house. Let the soil gas concentration remain constant at C₀. If diffusion is the only transport process, then the steady-state solution to (3) is taken as the concentration profile C(x) in the slab. The boundary conditions are:

$$\begin{aligned} C(L) &= 0 \\ C(0) &= C_0 \end{aligned} \quad (4)$$

The derivative $\partial C / \partial t$ is set to zero and (3) is solved to yield the profile of the interstitial concentration:

$$C(x) = \frac{C_0 \sinh\{(L-x)/\ell\}}{\sinh(L/\ell)} \quad (5)$$

Fick's law may be used to relate the gradient at x = L to the flux per area through the top of the slab. The total emanation rate from an

area A on the surface of the slab equals the flux of the interstitial concentration through the unoccluded area, ϵA . (The fraction of unoccluded area is easily shown to equal the porosity.) Therefore:

$$J = -\epsilon AD \left\{ \frac{\partial C(x)}{\partial x} \right\}_{x=L}$$

$$= \frac{A \lambda \epsilon \lambda C_0}{\sinh(L/\lambda)} \quad (6)$$

If $L/\lambda < 0.5$, then $\sinh(L/\lambda) = L/\lambda$ within 5% and (6) reduces to:

$$J = \epsilon A C_0 \lambda^2 / L \quad (7)$$

The boundary conditions given by (4) may be applied to a sample in the laboratory and the emanation measured. To calculate the flux from the surface of a slab of arbitrary thickness, ϵ and λ must be determined independently so that (6) may be used. From (7) it is evident that steady-state measurements on thin slabs will never separate ϵ and λ ; a measurement on a thick slab ($L > \lambda$) or a time-varying measurement on a thin slab is required. In laboratory measurements, a thin slab is preferred, since more radon can be collected over a shorter time interval, decreasing the time needed to complete the measurement. Furthermore, the area of the sample is limited to the disk that may be cut from a standard 6 inch (15 cm) diameter test cylinder. For an area this size, edge effects render one-dimensional diffusion a poor approximation for thicknesses much in excess of 3 cm.

In this experiment, a concentration C_0 is applied to one surface of a thin slab in a step function at $t=0$. The subsequent time-varying concentration profile is provided by the time-dependent solution to (3). This equation may be simplified by choosing a sufficiently thin sample, in which case steady-state is reached rapidly enough to justify dropping the decay term, C/ℓ^2 . This yields the heat equation:

$$\partial^2 C / \partial x^2 = \frac{1}{D} \partial C / \partial t \quad (8)$$

The appropriate boundary conditions and initial conditions are:

$$\begin{aligned} C(x=0, t>0) &= C_0 \\ C(x, t \leq 0) &= 0 \\ C(x=L, t) &= 0 \end{aligned} \quad (9)$$

The solution to (8) gives the interstitial radon concentration within the slab:

$$C(x,t) = C_0 \{1-x/L\} - \frac{2C_0}{\pi} \sum_{n=1}^{\infty} \frac{1}{n} \sin\left(\frac{n\pi x}{L}\right) e^{-Dn^2\pi^2 t/L^2} \quad (10)$$

Fick's Law is applied to (10) to determine the total accumulated activity I from an area A during a time interval T beginning at $t=0$:

$$\begin{aligned} I &= -\epsilon A \int_0^T D \left\{ \partial C(x,t) / \partial x \right\}_{x=L} dt \\ &= J \left\{ T + \frac{2L^2}{D\pi^2} \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} (1 - e^{-Dn^2\pi T/L^2}) \right\} \end{aligned} \quad (11)$$

where $J = \epsilon ADC/L$ is the steady-state flux from A. For large T, (11) simplifies to:

$$I = J \left\{ T - \frac{L^2}{6D} \right\} \quad (12)$$

This approximation is accurate to better than one percent for $T > L^2/2D$.

To characterize a sample, I is measured for some $T > L^2/2D$, and J is measured after the system has attained steady-state. $D = \lambda \ell^2$ is calculated by hand from (12). D is checked by calculating I from (11) with a computer. If the measured and calculated values of I agree closely, ϵ is determined from (7). D and ϵ may then be used with (11) to predict the emanation for shorter time intervals and different thicknesses. This over-determination provides a check for the theory.

EXPERIMENTAL METHOD

The concrete samples examined were 2-3-cm thick, 6-in. diameter disks taken from test cylinders. These were studied using the experimental apparatus illustrated in Figure 1. Aluminum cans 2.5-cm deep are sealed to each surface of a sample with a bead of silicon sealant. One can serves as a source and the other as a collector for radon emerging from the opposite side. The source can is the same diameter as the sample so that the full surface of the sample is exposed to the concentration maintained inside, about 7000 pCi/l. The collection can is 9.5 cm in diameter and is centered on the opposite surface of the disk.

Each can has two ports connected by surgical tubing to the inlet and outlet valves of a modified 12.5-liter compressed-air paint can so that each of the concrete surfaces is part of a closed loop. A polyvinyl fluoride bag filled with approximately 0.25 liters of air is attached to each loop so that both sides of the sample remain at ambient pressure. The paint can on the source loop is filled with air containing approximately 90 nanocuries of radon at the start of a measurement. Over the five-hour time period of a measurement, the concentration remains constant to within 5%, despite losses from decay and diffusion into the concrete disk. A peristaltic pump continually moves the air at 0.1 l/m to maintain a stable concentration in the source loop. A second peristaltic pump on the collection loop moves the air at 0.5 l/m to carry air away from the sample and dilute it with air in the paint can, maintaining a radon concentration close to zero in the collection can.

Each run measures the increase in emanation rate. The measurement sequence is divided into several contiguous time intervals beginning at $t=0$ and extending past the time required to attain steady-state diffusion. At the end of each interval, the paint can in the collection loop is replaced. The air in the paint can is flushed with helium at 0.8 l/m through a trap filled with glass wool and cooled with liquid nitrogen, at which temperature the radon condenses. The trap is then evacuated and sealed, after which it is allowed to warm to room temperature. Helium is then admitted into the trap as a carrier gas and pumped into an evacuated scintillation flask. This transfer procedure has been reproducible within 5% (In80b). After permitting the daughters to reach equilibrium, the alpha activity in the scintillation flask is counted by a photomultiplier tube. The amount of radon accumulated in the can is

given by:

$$I_0 = \frac{\lambda N}{6.66q(e^{-\lambda t_w})(1-e^{-\lambda t_c})} \quad (13)$$

where λ is specified in minutes and:

N is the number of counts during the count time,

t_c is the count time (min),

t_w is the time interval between the midpoint

of the collection interval and the start

of the count (min),

q is the alpha counting efficiency of the scintillation

flask and photomultiplier system, and

I_0 is the activity in pCi in the can at the

end of the collection period.

The background attributable to ambient radon in the air sealed in the cans is determined by transferring one can immediately as a blank. At the end of a measurement, the source concentration is sampled by opening an evacuated scintillation flask to the source loop. This concentration is used to normalize the time-varying and steady-state measurements to a source concentration of 10,000 pCi/l.

EXPERIMENTAL RESULTS

Three samples were tested. Disks of 2-cm and 3-cm thickness were cut from a test cylinder poured in Chicago, Illinois. A 2.5-cm thick disk was taken from a cylinder poured in Salt Lake City, Utah. The density of the first two samples was 2.40 g/cm^3 , with aggregate typically less than 1 cm in diameter. The density of the 2.5-cm sample was 2.25 g/cm^3 , with aggregate typically 1-1.5 cm in diameter. The edges of each disk were coated with approximately 0.5 g/in^2 of a two part epoxy sealant to assist in forcing a one-dimensional diffusion profile. Earlier tests performed by our group have shown this sealant to be unreliable in completely forcing one-dimensional diffusion; the geometry of the experiment is designed to avoid sole reliance upon any coating.

The integrated flux of radon from the 2-cm sample was measured over four contiguous thirty-minute intervals beginning at $t=0$. This buildup measurement was repeated five times. The steady-state flux was determined seven times. The source concentration was sampled twice at the end of each run, and for analysis purposes, the data were normalized to a constant source concentration of 10,000 pCi/l. The average activity accumulated after two hours and the average steady-state flux yielded ε and ℓ as 0.068 ± 0.007 and $12.6 \pm 0.3 \text{ cm}$, respectively. Equation (11) was used with ε and ℓ to calculate the activity expected to be collected during each thirty-minute interval. Figure 2a summarizes the experimental and calculated activities. The average normalized activity collected during each time interval is displayed versus the time elapsed at the end of that interval. The activity calculated is also shown. Equation (11) was also used with ε and ℓ to generate the solid curve that

predicts the accumulated activity, I , versus time. The average activities accumulated after 0.5, 1.0, and 1.5 hours, plotted as open circles, are in good agreement with the curve. The average activity accumulated after 2.0 hours will, of course, fall on the curve because it was used to determine ϵ and ℓ .

The values of ϵ and ℓ found from the 2-cm sample were used to predict the activity collected from the 3-cm sample during five contiguous one-hour intervals beginning at $t=0$. These activities were measured nine times. The gross flux rose significantly with each measurement until, by the third run, it had leveled off near the predicted values. The first two runs were performed within one week after attaching the sample to the source and collection cans with silicon sealant. The first two runs are shown in Figure 2b. The cause for the transient rise in flux is not known. The other samples were tested at least one week after they were sealed to the apparatus, and no transient effects were observed.

The averages of the last seven runs on the 3-cm sample are shown in Figure 2b. Both theoretical curves are generated from (11), using the values of ϵ and ℓ obtained from the 2-cm sample. The excellent agreement between experiment and theory lends strong support to the assumption that concrete may be treated as a homogeneous diffusion medium for radon. The flux during the fifth hour may be taken as the steady-state value. The steady-state flux and the average activity accumulated after four hours yield ϵ and ℓ as 0.069 ± 0.005 and 12.5 ± 0.2 cm.

The 2.5-cm sample, from the second test cylinder, was tested over four contiguous one-hour intervals beginning at $t=0$. Two runs were

performed. The average integrated flux during the fourth hour is taken as the steady-state value. The average activity accumulated after four hours and the steady-state flux determined ϵ and ℓ as 0.32 and 16.9 cm. The results are shown in Figure 2c. The individual results from both runs are plotted against the predictions for the activity accumulated during each interval. The averages of the activity accumulated after 1, 2, and 3 hours are plotted against the solid curve generated from (11).

CONCLUSIONS

The diffusion of radon through two different types of concrete was successfully modeled by two diffusion parameters determined experimentally with a time-dependent method. This result lends strong support to the hypothesis that concrete may be treated as a homogeneous diffusion medium for radon. In addition, the time-dependent method has proved to be a more rapid and convenient procedure for measuring diffusion parameters than previous methods reported in the literature.

The first concrete tested yielded a porosity of 0.068 and a diffusion length of 12.6 cm ($D = 3.34 \cdot 10^{-4}$ cm²/sec). The respective values for the second concrete were 0.32 and 16.9 cm ($D = 6.01 \cdot 10^{-4}$ cm²/sec). It is not known how representative either of these pairs of values is for concrete. With this qualification, the radon concentration can be calculated inside a tight one-story house due to diffusion of soil-gas radon through a 10 cm thick foundation slab composed of either material. If the ventilation rate is 0.1 air changes per hour and the soil gas diffusive concentration is 1000 pCi/l, the soil-gas contribution to the indoor concentration is 0.24 pCi/l for the first material

and 2.1 pCi/l for the second.

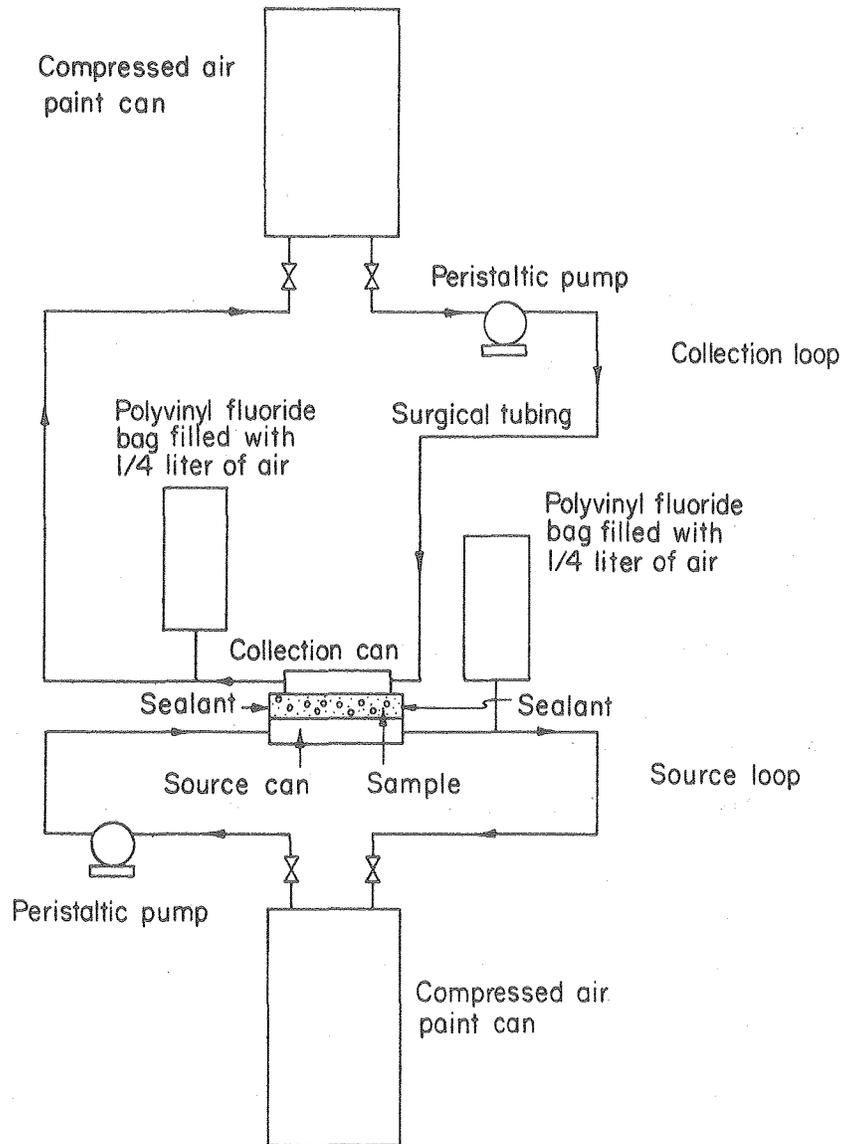
It is suspected in many cases that diffusion through the slab is not the primary source of radon indoors; radon may enter from transport through cracks or openings in the concrete. Furthermore, the driving force may be a pressure differential rather than a concentration gradient. Nonetheless, knowledge of the diffusion characteristics of concrete does indicate the extent to which diffusion contributes to indoor radon concentrations, or, alternatively, the potential for concrete to act as a barrier to radon from soil gas.

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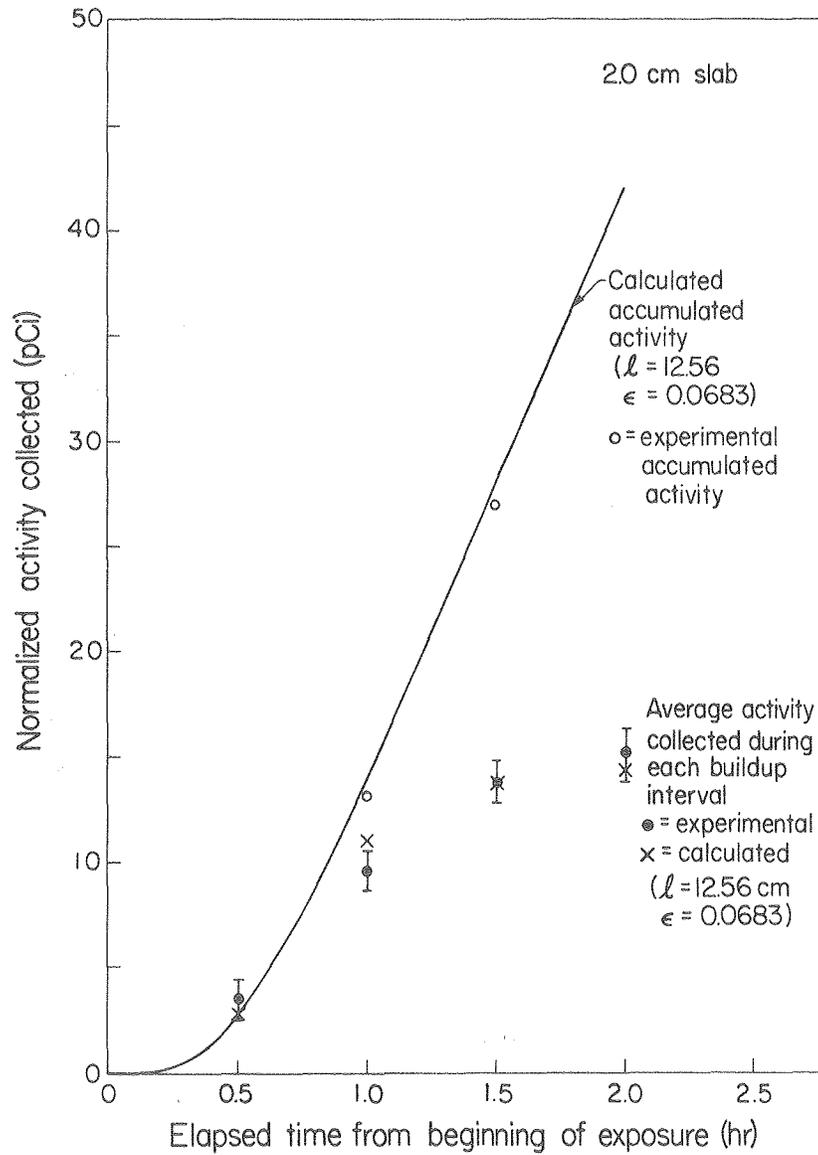
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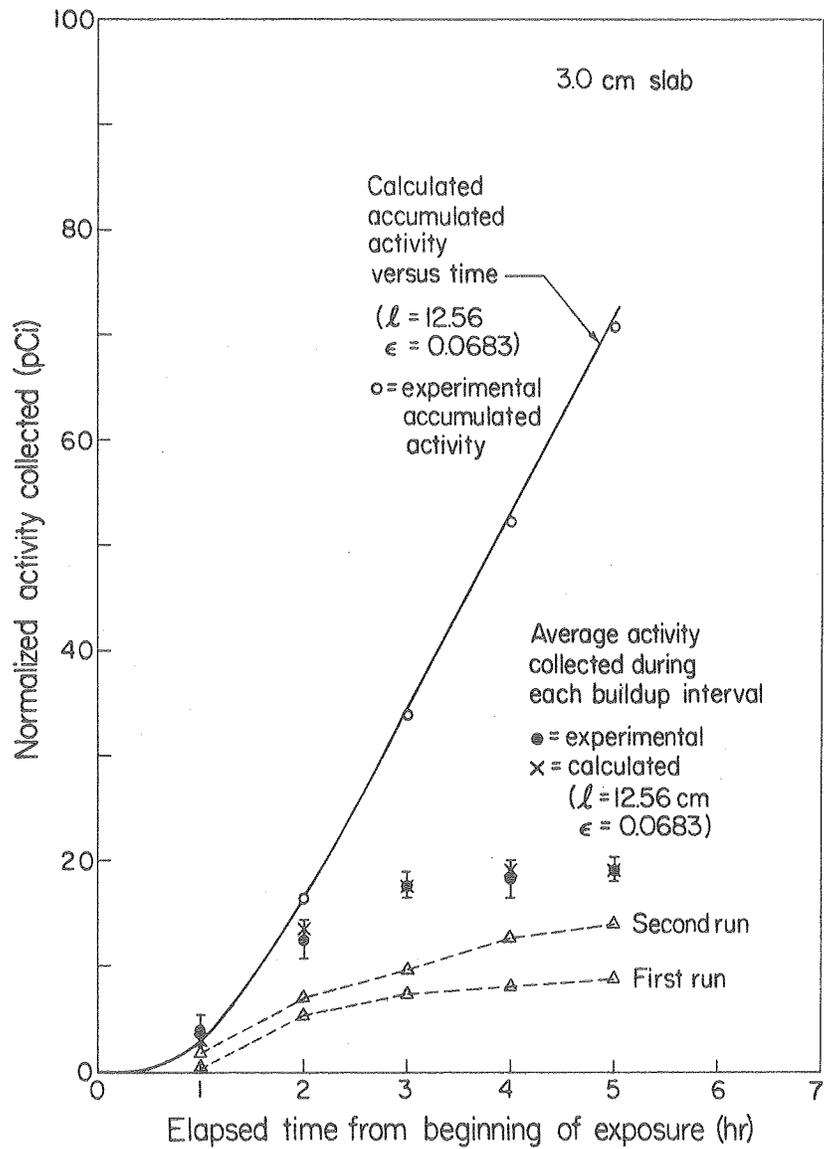
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Fig. 1: Apparatus used to measure the diffusion of radon through concrete. The source loop exposes one surface of the sample to a constant concentration of radon beginning at $t=0$. The radon that diffuses out the opposite surface into the collection loop is transferred from the paint can to a scintillation flask.



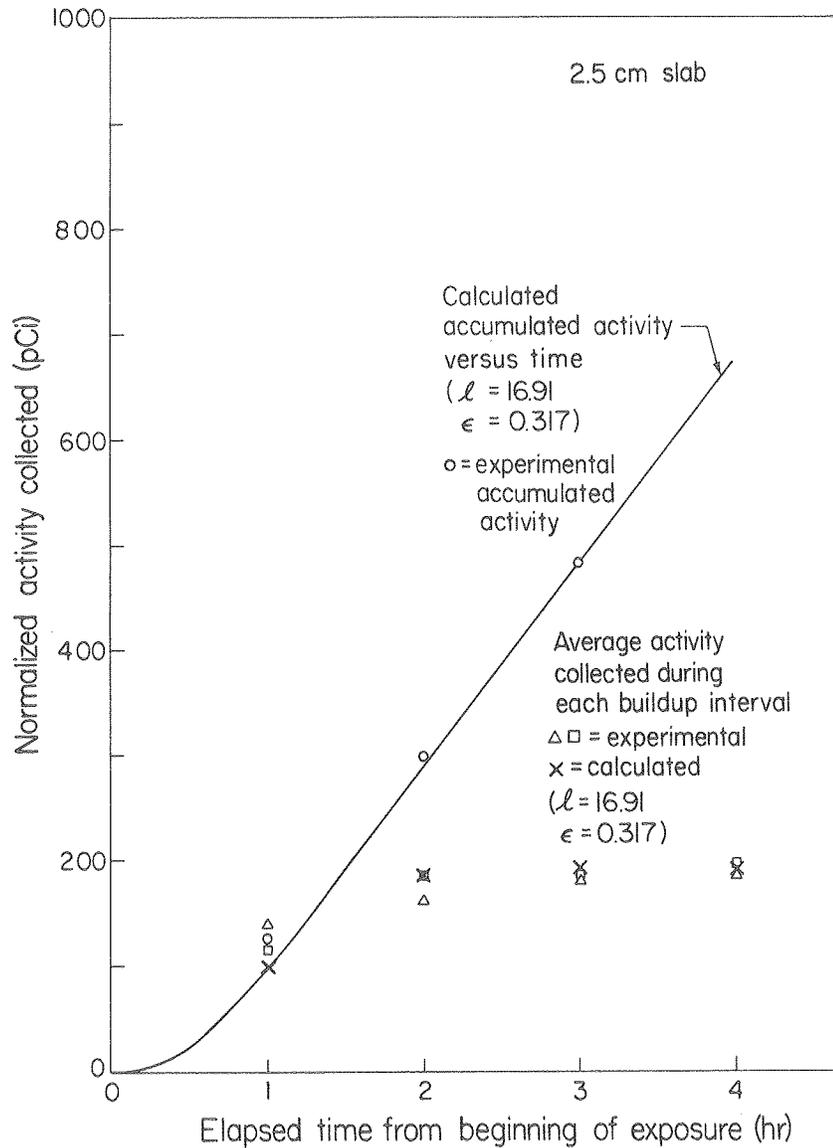
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Fig. 2a: Results for the 2-cm sample. The solid curve is generated from equation (12) and the diffusion parameters. Error bars indicate the standard deviation from the average experimental value.



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Fig. 2b: Results from the 3-cm sample taken from the same test cylinder as the 2-cm sample in Fig. 2a. The results of each of the first two runs and the averages of the last seven runs are shown. The theoretical curve is generated from the diffusion parameters found for the 2-cm sample.



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Fig. 2c: Results from a 2.5-cm sample taken from a different type of concrete.

