

MATHEMATICAL MODELING OF TIME DEPENDENT RADON FLUX PROBLEMS

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ABSTRACT

Background is given on the various equations used to describe diffusion and convective flow of radon in porous media. Bureau-developed equations for modeling diffusion and Darcy flow through multilayered porous media are described briefly, and examples of their use are given. Analysis results discussed include: The transient effect of pressure changes in different length models, time dependent flux changes in multilayered models, and radon flux through pinholes in sealants.

INTRODUCTION

Radon was discovered in 1900 by Friedrich Ernst Dorn, a German chemist, just 2 years after the Curies announced their discovery of radium. By 1905 researchers were measuring the radon concentration of soil gas and had noticed its variability as a function of atmospheric conditions¹. Since that time there has been a continual stream of reports and papers on radon. One author² estimated that over 1,000 papers have been written on radon or its daughters, and Tanner³ estimates that he has over 2,000 citations. Two excellent reviews of the radon literature are given by Tanner^{4,5}. He summarized many of the factors controlling radon flow, from diffusion through the mineral grain in which the parent radium is embedded, to radon's escape through open fractures.

Many variables affect the migration of radon: Decay rate of the isotope, gas diffusion coefficient in pore-filling fluid, porosity of the medium, velocity of the fluid, composition of the fluid, and, if the pore-filling fluid has more than one phase, the temperature dependent distribution of the radon among the phases. Tanner⁴ also provides three sets of characteristics to be considered for radon migration: "(1) Rock or soil unsaturated by liquids and free of open fractures or channels, (2) fractured rock or disturbed soil, unsaturated by liquids, and (3) saturated rock or soil." In this paper we are concerned with radon flow in only the first set of characteristics, within which radon can move by either diffusion or transport. Diffusion takes place when the radon isotope moves through the fluid filling the pores of the medium. Transport, also called convection, occurs when the fluid itself moves through the porous medium and carries the radon with it.

Many equations have been developed to describe the flow of radon in a porous medium (see the following discussion). However, most of them evaluate one-

dimensional planer type problems with steady-state diffusion or constant convection flow. None is applicable for evaluating radon flow in the conditions found in mines, especially the changes in barometric pressure that are encountered. Also, existing equations do not permit examining such radon control measures as overpressurization, cyclic pressurization, or radon barrier coatings. To satisfy this need for a better evaluation tool, a set of equations and computer codes have been developed for multilayered porous media radon emanation problems with changing barometric pressures. The general characteristics, capabilities, and limitations of our analysis are given following the discussions of some equations found in the literature. Example analyses of constant pressure changes, cyclic pressure changes, and pinholes in a radon barrier coating are described.

SIMPLE EQUATIONS FROM THE LITERATURE

The literature surveyed contains a number of solutions to various problems of radioactive gas movement through porous media⁴. One of the simplest expressions for evaluating the movement of radon from a planer source through a nonradioactive soil is given by

$$C = C_0 \exp \left(- \sqrt{\frac{\lambda \phi}{D}} z \right), \quad (1)$$

where C = concentration at distance z from the plane source,

C_0 = the concentration in the plane source,

λ = the radioactive gas decay constant,

D = the diffusion coefficient, and

ϕ = porosity of solid matrix (void volume/total volume).

The diffusion length is $\sqrt{D/\lambda\phi}$ and a 100-fold decrease in concentration will be found at a distance $z = 4.6 \times \sqrt{D/\lambda\phi}$.

A steady component of transport or convection in the direction of the diffusion was added to the equations by Grammakov in 1936 (as quoted by Tanner)⁴. If the transport component, v , is added to the previous equation, the one-dimensional solution is

$$C = C_0 \exp \left\{ \left(\frac{v\phi}{2D} - \sqrt{\frac{v^2\phi^2}{4D^2} + \frac{\lambda\phi}{D}} \right) z \right\}. \quad (2)$$

Tanner quotes a number of additional references for radon diffusion with and without transport mechanism acting. Some account for a radioactive overburden, differences in porosity between the diffusion media,

and even corrections for absorption of radon by soil water.

Another form of the idealized equations are those dealing with diffusion of radon through a radioactive body such as an ore sandstone, tailings, or even soil. These equations have been published a number of places^{6,7,8} and are repeated here for completeness. It is assumed that Fick's first law of diffusion holds for radon diffusion through porous media. Therefore,

$$J = -D \frac{\partial C}{\partial z}, \quad (3)$$

where J = radon flux across the total surface (not just the exposed pores),
 D = radon diffusion coefficient, and
 $\partial C / \partial z$ = the radon concentration gradient.

Using a steady state form of Fick's second law of diffusion and adding the radon production and decay terms, we have the following:

$$\frac{\partial C}{\partial t} = 0 = \frac{D}{\phi} \frac{\partial^2 C}{\partial z^2} - \lambda C + q, \quad (4)$$

where D = diffusion coefficient,
 ϕ = porosity,
 C = concentration of diffusing radon per unit volume of bulk medium,
 z = depth (positive inward from surface),
 λ = radon decay constant, and
 q = production rate for diffusing radon.

Solving equation (4) for C yields

$$C = (C_0 - C_1) \exp \left(-z \sqrt{\frac{\lambda \phi}{D}} \right) + C_1, \quad (5)$$

where C_0 = radon concentration at $z = 0$ and
 C_1 = radon concentration at depth ($z = \infty$) = q/λ .

Generally C_0 is very small compared with C_1 . For example, Schroeder et al.⁹ reported interstitial radon concentrations of 6×10^5 picocuries per liter (pCi l^{-1}). (The bulk concentration is about $1.2 \times 10^5 \text{ pCi l}^{-1}$.) In uranium mines the desirable airborne radon concentration is below 100 pCi l^{-1} . Therefore, for most situations C_0 can be set equal to 0, and equation (5) reduces to

$$C = C_1 \left\{ 1 - \exp \left(-z \sqrt{\frac{\lambda \phi}{D}} \right) \right\}. \quad (6)$$

Substituting (6) into equation (3) and evaluating the result at the plane $z = 0$ gives the radon flux at the air/porous media interface:

$$J = -\sqrt{\lambda D \phi} C_1 = -q \sqrt{\frac{D \phi}{\lambda}}. \quad (7)$$

Several other idealized problems have been solved for steady-state diffusion, as summarized by Tanner, p. 170⁴; "Koenigsberger (1928) and Bulashevich (1946, 1947) derived expressions for the concentration of radon-222 or radon-220 at the surface in alluvium covering a linear source. Khaykovich (1961) considers the distribution of radon migrating through an ore layer into a cylindrical borehole. Grammakov (1961a) gives solutions for several problems in spherical and cylindrical geometry. Solutions for various source

distributions have been summarized in the Soviet prospecting manual (Alekseyev, 1957, Sec 49)."

One equation developed more recently is that by Schroeder and Evans¹⁰, which describes radon diffusion in multilayered systems. These one-dimensional equations provide for the steady-state generation and diffusion of radon through any number of porous layers with or without significant quantities of radon generation in each layer. The porosity from layer to layer can be varied, but apparently the diffusion coefficient is assumed to be the same in each layer. These equations have been used to evaluate a number of situations such as radon migrating from an ore body through an essentially barren overburden, or the radon from a finite-sized layer of radium-rich sand escaping through an essentially barren soil to the atmosphere. These equations were also used^{11,12} to estimate the radon loss through a porous radon barrier material.

NONSTEADY-STATE MODELS

Steady-state conditions have received considerable attention, but transient conditions have received little. Measurements have been made of radon flux variation with wind, rain, barometric pressure, and freezing temperatures. Equations also have been derived empirically¹³ to describe some conditions in mines during constant- or rising-barometer situations. In 1974, Clements¹⁴, as part of his doctoral thesis, derived the first equations that combined molecular diffusion and transport (Darcy flow) components of the radon migration problem into a single analysis. He linearized the one-dimensional equations and used them to demonstrate the effect of a linearly rising or falling barometric pressure on the radon flux at the air/soil interface. A modification of Clements' work was used by Jeter et al.¹⁵ to calculate the amount of radon-222 and krypton-85 migrating from a constant radiation source (uranium ore body) through a nonradioactive overburden to the atmosphere. A variety of linear and sinusoidal barometric pressure changes were imposed at the surface to show the transient effect of a falling or rising barometer. In Jeter's work, it was found that barometric pumping did not enhance the detectability of a uranium ore body.

While these analyses, both empirical and theoretical, have advanced the capability of evaluating questions connected with radon emanation, they are not sufficient for analyzing the features found in uranium mines. Uranium ore bodies are finite, some are truncated by impermeable shale bands, and others grade off into nonmineralized sandstone. Mine surfaces are very irregular and on occasion are covered or strengthened with concrete or shotcrete. The pressure in a uranium mine is unsteady, with mine-caused pressure variations superimposed on the natural barometric pressure variations. Mine-caused pressure variations include the turning on and off of ventilation fans, opening and closing air doors, blasting pressures, and the movement of vehicles in the shaft or haulageways. Overpressurization of mines has been proposed as a radon control method⁹. Another control measure, polymeric radon barriers, has been proposed and demonstrated^{16,17,18,19} as effective for decreasing the emanation of radon into a mine opening. However, a question has been raised about the effect

of a pinhole in such a barrier coating^{11,12,16,20}. Since it was not possible to attack any of these problems using the equations available in the literature, a small effort was undertaken within the Federal Bureau of Mines to develop equations that would provide answers to some of these problems. These equations, which are very complex, are being reported elsewhere²¹, but the model characteristics are given in the following section.

MATHEMATICAL MODEL

The two-dimensional, cylindrical coordinate equations developed by the authors permit us to evaluate a multilayered system with an arbitrarily varying barometric pressure. With these equations, we can examine many different kinds of radon control problems. The present computer code is set to analyze radon (Rn 222), but with a simple change of constants, it can be used to evaluate problems associated with thoron (Rn 220) or any other radioactive gas in a porous medium. The general characteristics of the mathematical model are given in figure 1. This model

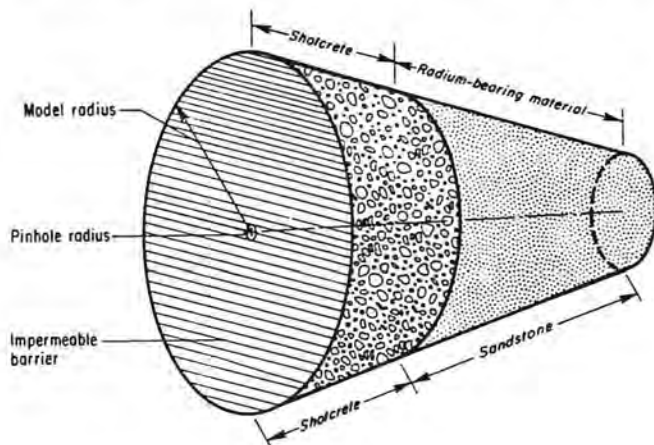


FIGURE 1. Idealized view of cylindrical mathematical model.

may be visualized as a cylindrical core cut perpendicular to the mine surface. The three zones of material include an ore zone (which contains a radiation source term), a nonradioactive zone (like shotcrete or concrete lining), and a flawed radon barrier coating. In our case the flaw is a pinhole with a definable radius. The polymeric coating is treated as an impermeable barrier intimately attached to the porous media (sandstone or shotcrete). This models the spray application used in mines by Franklin^{18,19} where a penetrating base coat is applied to the sandstone or shotcrete. This coating is then covered by a thicker (0.25- to 0.50-mm) top coat. One or more layers may be left out of a particular analysis. If a preformed film is loosely attached to the rock, this mathematical approach is not applicable since a provision for an air gap would have to be added.

A finite difference mathematical approach is used to evaluate the equations. This type of analysis requires that a grid be defined giving the dimensions for the two porous media.

For each of the porous media, the thickness, porosity, permeability, and diffusion coefficients may be defined independently. In addition, the air pressure on the face of the model can be held constant or varied in any arbitrary fashion. As mentioned, the size of the pinhole in the impermeable barrier may be defined, or the barrier may be left off entirely. Since this is a time-dependent analysis, a time step must also be specified. The magnitude of the time step is controlled to some degree by the analysis desired (diffusion only or diffusion and Darcy flow), the size of the geometric grid step, and the physical properties.

This mathematical model has been used to examine a number of radon emanation questions, and the following paragraphs summarize the results of some of these analyses and point out their implications to radon control problems.

PRESSURE VARIATION EFFECT CHANGES

Changes of radon flux in response to changes in barometric pressure have been reported throughout the literature^{6,13,14,16,18,22}. In fact, as early as 1905 Brandes, as reported by Joly and Smyth¹, noticed changes in soil gas radon concentration in response to atmospheric conditions. Diurnal changes in barometric pressure are responsible for corresponding changes in the radon flux or radon concentrations. Overpressurization of uranium mines has been discussed as one way to inhibit the emanation of radon into the mine workings⁹. In discussing overpressurization of mine workings, two situations must be considered: (1) the infinite sink, which permits a continuous convective airflow away from the pressurized area, and (2) the finite-sized porous body, which will ultimately become entirely filled with the higher pressure. The computer code can handle either case, but only finite ore body situations are discussed here.

Analyses were performed to examine the effect of pressure changes on several model (ore body) thicknesses, (6.75 to 135m), without shotcrete or a barrier coating. The porosity, permeability, and diffusion coefficient values used are 0.5, $1 \times 10^{-8} \text{ cm}^2 \text{ s}^{-1}$, and $0.022 \text{ cm}^2 \text{ s}^{-1}$, respectively. The initial radon concentration in the porous media followed equation (6). Figure 2, for a 6.75m ore body, clearly shows that the effect of a fixed pressure change is a transient phenomenon. For this model, the Darcy flow is 75 percent complete within 45 minutes and 100 percent complete in 3 hours. The peak change in emanation occurs approximately 1 hour after the onset of the pressure change. In 20 to 30 hours from the start, the radon flux approaches the original steady state condition.

Alternate negative 4-percent and positive 4-percent pressures were imposed at 12-hour intervals on the 6.75m ore body. For this finite ore body, each imposition of a pressure change was characterized by a peaking within about 1 hour (figure 3) and then a falling off just as in the curves in figure 2. The magnitude of the peaks was essentially the same when the model was subjected to eight cycles (8 days). This indicates that the radon in this size reservoir is not being depleted by this barometric pumping. The radon concentration gradient within the porous medium is altered by the pressure changes. The greatest change in the gradient, figure 4, corresponds to the peaks in flux, when contaminated air is being pulled

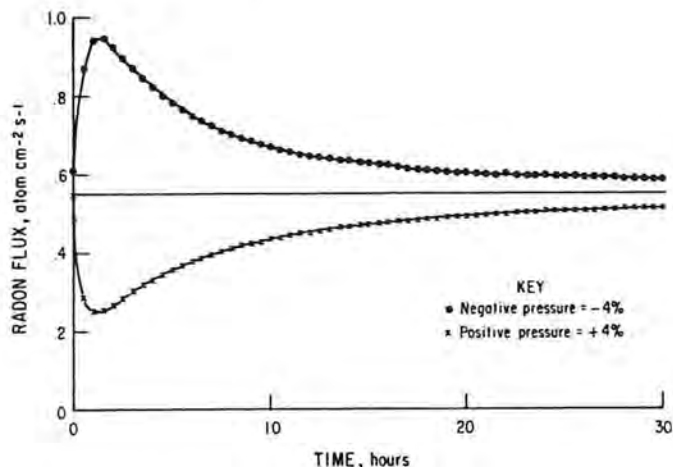


FIGURE 2. Effect of constant 4-percent over or under pressurization on radon flux, 6.75m radioactive body.

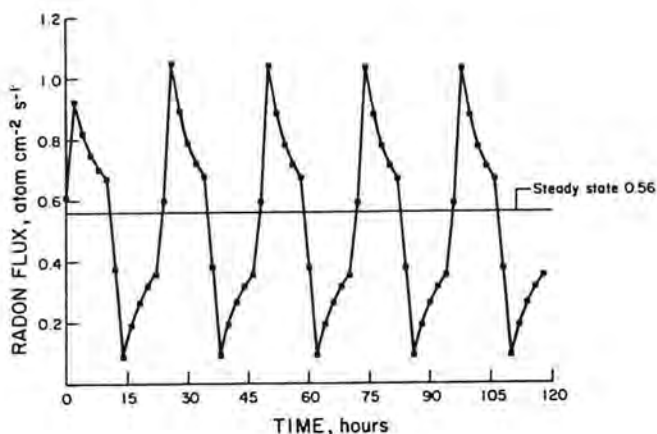


FIGURE 3. Radon flux from cyclic 4-percent under and over pressurization, 6.75m porous body.

out of the porous body or clean air being forced into it. [The radon concentration within the porous medium is normalized by q/λ to give values between 0 and 1, $\lambda = 2.1 \times 10^{-6} \text{ s}^{-1}$ and for figures 2-7, $q = 0.28522 \times 10^{-23} \text{ gm cm}^{-3} \text{ s}^{-1}$.]

The largest finite ore body model analyzed to date is 135m. A continuous 4 percent pressure change, negative and positive, caused a rise or drop in radon flux that is larger but slower, figure 5, than that of 6.75m ore body. The peaks occur at 10 hours after the onset of the pressure change and Darcy flow is not complete for several hundred hours. The flux approaches the steady-state condition some time beyond 400 hours. Alternate 4-percent negative and positive pressures were also imposed on this model. In the first run of this analysis we found that the pressure

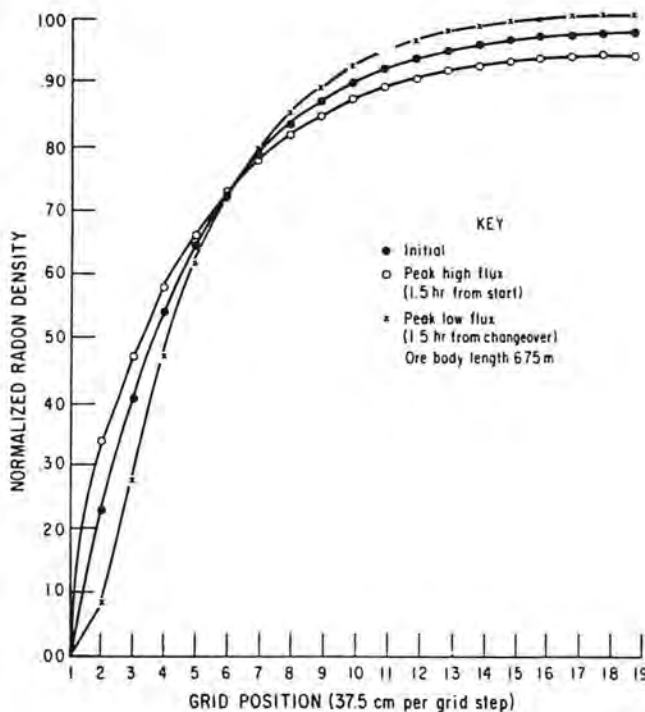


FIGURE 4. Normalized radon density in a 6.75m porous body at peak flux with a 4-percent overpressure and 4-percent underpressure. Steady-state radon density given for reference.

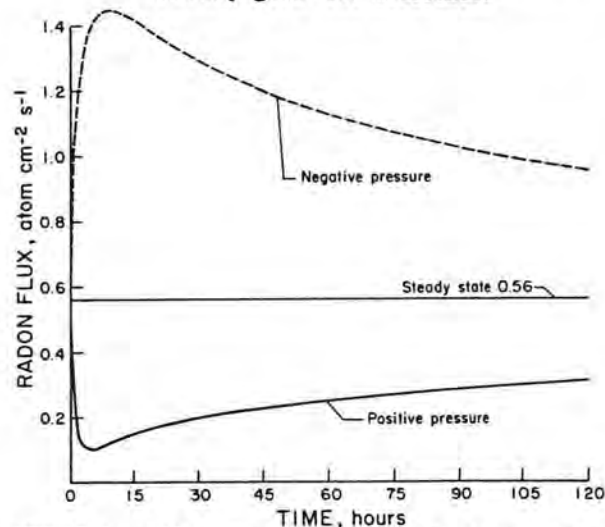


FIGURE 5. Effect of constant 4-percent over and under pressurization on radon flux, 135m radioactive body.

could not be changed in a single time step. At the 12th hour air was still moving across the porous media/air interface, and an attempt to reverse the flow instantly caused a mathematical instability. This was overcome by changing the pressure in several increments. The flux cycles in figure 6 are truncated versions of the figure 5 curves, and except for a large flux for the first cycle all of the peaks are nearly equal. So even on this large model barometric pumping doesn't change the average flux. Figure 7 shows the radon concentration gradient as initialized,

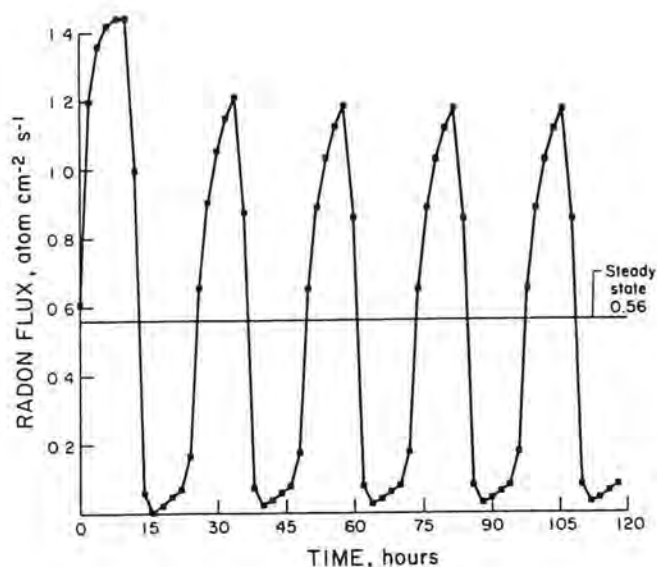


FIGURE 6. Radon flux from cyclic 4-percent under and over pressurization 135m porous body.

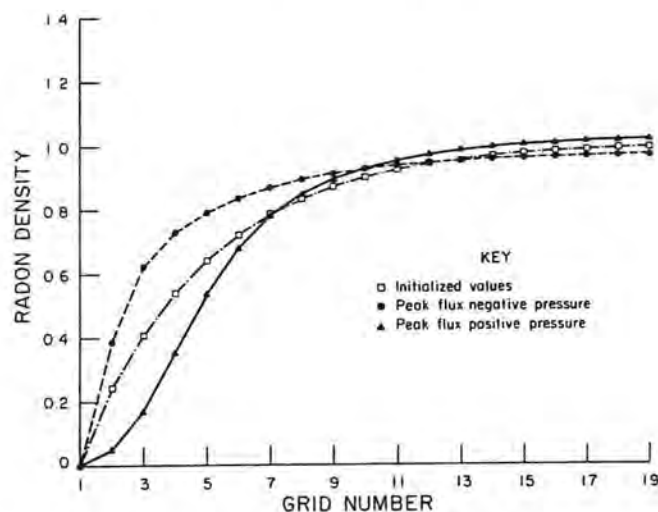


FIGURE 7. Normalized radon concentration gradient in a 135m porous body at peak flux.

first negative cycle and first positive cycle. The concentration gradient changes more than it does for the 6.75m ore body.

RADON BARRIER COATING PINHOLE ANALYSES

The influence of pinholes in a radon barrier coating has been discussed several times in the fairly recent literature^{11,12,20}. However, in these discussions, the influence of pinholes in the coating was not treated rigorously, and at best it was treated using statistical methods¹⁶. In our studies of pinholes, several different model configurations were used. In one, the radon barrier coating was placed

directly on the radioactive porous medium and the pinhole of some arbitrary size was defined in the barrier coating. This case is analogous to the Dakota mine experiments performed by the Federal Bureau of

Mines^{16,17,18}. Another form of the model used all of the features available in the code and defined a 5-centimeter-thick shotcrete layer between the radon barrier material and the radioactive sandstone (material properties are given in table 1). This simu-

TABLE 1. Properties used in two-layer analysis

Item	Shotcrete ¹	Sandstone ²
Porosity	0.05	0.50
Permeability, cm ²	1×10^{-10}	1×10^{-8}
Diffusion coefficient, cm ² s ⁻¹	1.69×10^{-5}	0.022
Radon source term gm cm ⁻³ s ⁻¹	—	0.24447×10^{-23}

¹Values taken from Culot et al.²⁴, concrete basement wall.

²Values taken from Clements¹⁴, Socorro, N. Mex. soil.

lates the situation such as at the Twilight mine¹⁹, where a layer of shotcrete was used to smooth out the irregular mine surface and fill large fractures. A further refinement on the multilayer model analyses was the manner in which the initial conditions were defined. In one case, the radon concentration gradient within the radioactive porous media was defined by the normal steady-state condition of equation (6). The shotcrete held no radon at $t = 0$, which simulates an unlikely condition in the field where the shotcrete and the radon barrier material are placed at the same time. The second case is a little more realistic, in that the two-layer steady-state radon distribution within the shotcrete and the sandstone was defined as the initial condition²³. The impermeable barrier with its pinhole is placed at $t = 0$.

The most distinctive feature in the results for the two-layer initialization is the rapidity with which the radon flux through the pinhole reaches a steady-state condition. The time required is approximately 30 to 50 hours for the two-layer initialization, compared with 700 to 800 hours for the other case, figure 8. This is also reflected in the radon concentration gradient within the porous media, figures 9 and 10. Because the concrete itself has such a low diffusion coefficient and porosity, it serves as a barrier to retard the loss of radon gas from the sandstone and thereby increases the radon concentration gradient in the sandstone. The two-layer initialized radon concentration at the shotcrete-sandstone interface is nearly 0.98, normalized, and it takes about 800 hours for the sandstone-only initialization to reach this interface concentration.

As mentioned previously, the real concern about the pinholes in radon barrier materials is whether they will release excessive quantities of radon and thereby destroy the barrier's effectiveness. Looking just at the flux coming from a single pinhole, figure 8, or even the actual atoms per second escaping from the pinhole, will tend to lead people astray because there

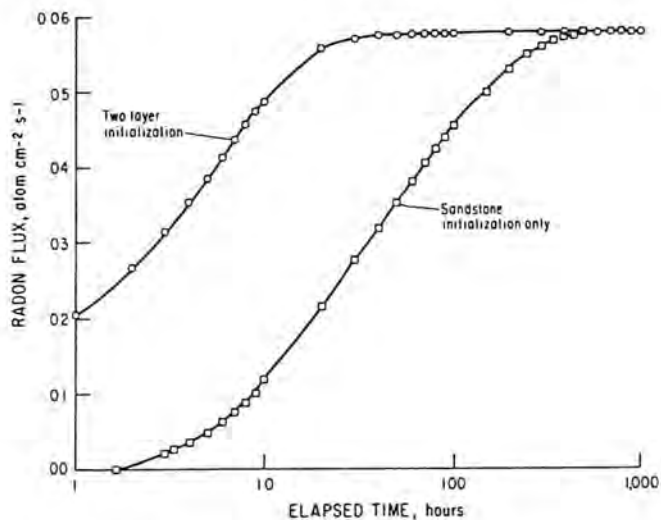


FIGURE 8. Radon flux through a 0.1-cm radius pinhole in an impermeable barrier covering 5-cm-thick shotcrete over a 11.5m radioactive porous body for one-layer and two-layer initialization.

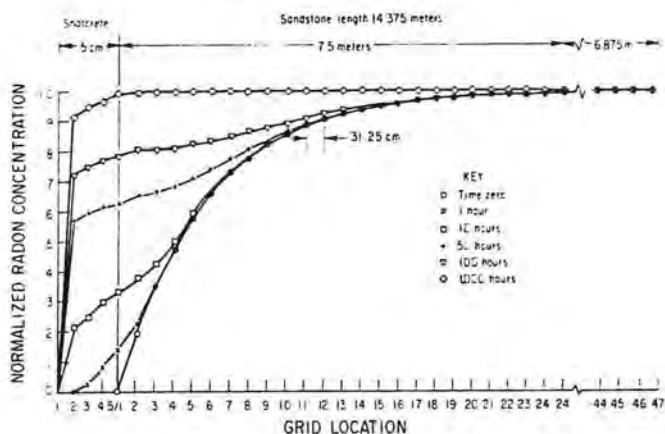


FIGURE 9. Normalized radon density for various times after start for 0.1-cm-radius pinhole, 5-cm shotcrete, and 11.5m radioactive body, using one-layer initialization.

is no solid base upon which to make comparisons of pinhole loss. Also it is likely that there will be more than one pinhole in a polymeric barrier coating. The question of many pinholes in a surface is not addressed specifically by the mathematical formulation. However, if there are many close pinholes, it appears logical that they will interact to deplete the reservoir, making the overall radon gradient in the porous medium less steep. This in turn will reduce the flux at each pinhole. Mathematically, the boundary at the midpoint between two pinholes is a noflow condition such as that already defined for the cylindrical walls of the model. The effect of close pinholes then could be approximated by setting the model radius equal to half the pinhole spacing. Some analyses were made with small radius models (less than 20 cm), and the pinhole flux was lowered. However, this was not a

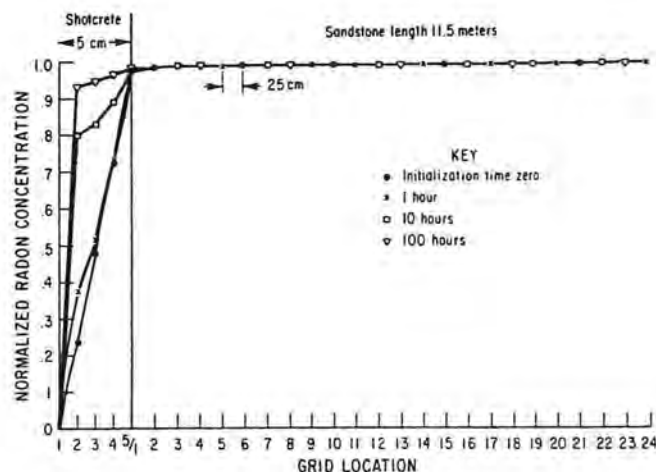


FIGURE 10. Normalized radon density for various times after start for 0.1-cm-radius pinhole, 5-cm shotcrete, and 11.5m radioactive porous body, using two-layer initialization.

truly rigorous three-dimensional analysis; therefore, the following conservative alternative was chosen.

Considering all factors, a conservative estimate of the radon flux from a multi-pinhole surface can be made by assuming that the maximum flux is available from each pinhole as though its neighbor was noninterfering. The actual loss of radon through a given pinhole density was calculated using the maximum flux, pinhole area, and number of pinholes per square meter. The resulting total escape was compared to the total flux for a 1-square-meter surface of 4,800 atoms per second. The results of this analysis using 1 to 10,000 pinholes per square meter are given in figures 11 and 12. Figure 11 gives the total loss through a

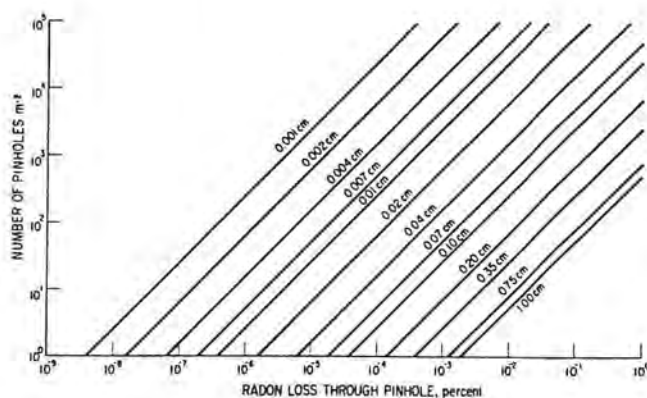


FIGURE 11. Percentage loss of radon through various number and size of pinholes in a barrier coating on shotcrete and sandstone. Numbers on lines indicate pinhole radius, cm.

combination of shotcrete and a pinhole-flawed barrier coating. One might argue that obviously the pinholes caused no difficulty in this system, because the shotcrete itself was given such good barrier properties.

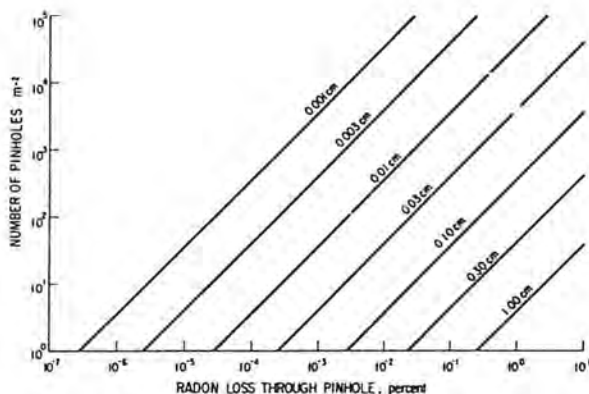


FIGURE 12. Effect of multiple pinholes in an impermeable barrier coating placed directly on a radioactive porous medium. Numbers on lines indicate pinhole radius, cm.

Figure 12, on the other hand, represents the case in which the impermeable barrier is placed directly on a very porous material. This is something of a worst-case analysis, since underground sandstones would not have a 50-percent porosity. Even in this case, thousands of visible-size pinholes per square meter would be required to cause a significant (10 percent) damage to the barrier efficiency. From these latter results, it appears that the extreme concern about pinholes in polymeric radon barriers is not justified.

SUMMARY

A background review of equations used to evaluate radon diffusion through a porous medium has been given. Examples of several calculations using the recently developed finite-difference code for diffusion and Darcy flow of radon through porous media have been given to illustrate the variety of problems that can be analyzed. If a steady pressure or vacuum is put on a finite-sized ore body, any initial decrease or increase in flux will gradually be overcome by the diffusion process that reestablishes the normal steady state condition. The results of these analyses indicate that barometric pumping is not significant in attenuating the amount of radon coming out of an ore body. The radon concentration gradient within the porous media is changed in response to the pressure cycling. The radon concentration gradient in a porous medium behind a barrier coating becomes very steep, and this increases the flux at the pinhole to six times the free open face flux. However, the actual loss of radon through the pinhole is considerably less than for an unprotected surface. Even a worst-case analysis given here indicates that thousands of visible pinholes would have to be scattered throughout the coating to cause a significant (10 percent) loss of barrier coating effectiveness.

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