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A Mathematical Model for Radon Diffusion in Earthen Materials

Prepared by K. K. Nielson, V. C. Rogers/RAE

Rogers & Associates Engineering Corporation

Pacific Northwest Laboratory

Prepared for
U.S. Nuclear Regulatory
Commission

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Prepared by
K. K. Nielson, V. C. Rogers, RAE

Rogers & Associates Engineering Corporation
P. O. Box 330
Salt Lake City, UT 84110

Under Subcontract to
Pacific Northwest Laboratory
Richland, WA 99352

Prepared for
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ABSTRACT

Radon migration in porous, earthen materials is characterized by diffusion in both the air and water components of the system as well as by the interaction of the radon between the air and water. The size distribution and configuration of the pore spaces and their moisture distributions are key parameters in determining the radon diffusion coefficient for the bulk material. A mathematical model is developed and presented for calculating radon diffusion coefficients solely from the moisture content and pore size distribution of a soil, reducing the need for resorting to radon diffusion measurements. The resulting diffusion coefficients increase with the median pore diameter of the soil and decrease with increasing widths of the pore size distribution. The calculated diffusion coefficients are suitable for use in simple homogeneous-medium diffusion expressions for predicting radon transport and compare well with measured diffusion coefficients and with empirical diffusion coefficient correlations.

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

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INTRODUCTION

Radon emissions from uranium mill tailings have long been recognized as a potential health hazard. Consequently, an important feature of any uranium mill tailings management program is the proper long-term stabilization of the tailings to adequately reduce radon emissions. The generally accepted means of achieving stabilization is to cover the tailings with earthen materials. Since radon is a chemically inert gas whose principal isotope, ^{222}Rn, has a radioactive half-life of 3.8 days, it can migrate considerable distances through earthen materials before decaying to a solid daughter product. The earthen cover must therefore be of sufficient thickness to contain the radon throughout several half-lives in order to minimize the fraction which ultimately escapes into the atmosphere. The thickness of cover required to reduce the flux of escaping radon to a given level can be calculated from the radon diffusion coefficient of the cover material.

Radon diffusion coefficients for earthen materials have traditionally been determined from laboratory measurements with the subject soil at a prescribed moisture content and compaction. However, it is often desirable to estimate the diffusion coefficient of materials under varying conditions for which measured values are not available. The Nuclear Regulatory Commission (NRC) has used a simple correlation of diffusion coefficient with moisture(1) to predict required earthen cover thicknesses in their Generic Environmental Impact Statement (GEIS) on Uranium Milling.(2) It is recognized, however, that moisture profiles are variable, and that soil compaction, texture, and other parameters also have a significant influence on diffusion coefficients. Therefore, it is desirable to develop a mathematical model to describe the radon diffusion coefficient from basic soil parameters rather than predicting it from empirical correlations. The present model was developed for NRC under a subcontract with Battelle

Pacific Northwest Laboratory to provide the necessary mathematical description of the radon diffusion process to accurately predict radon diffusion coefficients from basic soil parameters.

Previous Studies

Researchers have long been interested in the diffusion and transport of radon through porous materials. Early studies of radon in the natural environment(3-13) have been supplemented by research dealing specifically with the diffusion and transport of radon produced in uranium mill tailings(11,14-16) and ore minerals.(17,18) In particular, References 3 and 19 contain excellent reviews of the general topic, and an excellent survey of the early Russian literature is given in Reference 20. The early studies concerned with the diffusion of radon from uranium mill tailings were based on experiments with the diffusion of radon through tailings, soil and concrete. Measurements made during these experiments were compared with diffusion theory and from the comparison, diffusion coefficients were deduced. In other laboratory experiments, (1,21,22) the diffusion of radon through various tailings and cover materials was measured. Diffusion coefficients were deduced from both radon fluxes and from radon gas concentration profiles. Rapid transient methods for measuring radon diffusion coefficients have also been recently reported.(23,24)

Two recent field tests(25,26) using uranium tailings materials also have yielded information associated with the diffusion coefficient. In one test(25) surface radon fluxes were measured for various thicknesses of a cover material placed over a small plot of tailings. The diffusion coefficient obtained from a least-squares fit of the flux data was consistent with the laboratory measurement on similar material. In the other field measurement(26) the diffusion coefficient was deduced from in-situ borehole logging of the ^{226}Ra and ^{222}Rn concentrations in acidic and alkaline tailings. Additional field measurements are in progress for a variety of single and multilayer earthen and synthetic covers,(27,28) and laboratory measurements of diffusion coefficients in similar multilayer systems have also been reported.(24)

One common feature pervades the bulk of the literature on radon diffusion in earthen materials and that is the necessity of basing the determination of the diffusion coefficient upon experimental radon measurements. Although these measurements are usually time-consuming and cumbersome, nevertheless, they allow for the accommodation of other mechanisms and uncertainties not explicitly quantified in the diffusion theory formalism.

In order to predict the diffusion coefficient of radon from physical properties of an earthen material without conducting radon measurements, two simple correlations have been proposed. One of these is the correlation with moisture(1) which was used in the GEIS on uranium milling.(2) The other is a correlation with the air-filled porosity of the soil.(29) Similar functions of air-filled porosity have been used with other soil gases in agricultural applications.(30) Although these correlations permit estimation of diffusion coefficients from soil properties rather than diffusion measurements, their basis is still a series of measured values of diffusion coefficients.

Scope of Present Work

This report describes the mathematical framework and methodology for estimating radon diffusion coefficients without relying on fitted parameters to radon diffusion data. The formalism considers the detailed composition of the pore fluid as well as a statistical definition of the pore structure. The pore fluid is modeled as a two-phase mixture of air and water, with radon diffusion occurring in both phases, and with radon exchange occurring between the air and water. The pore structure is modeled from the measured pore size distribution of the soil, and is described by the weighted average of all combinations of single and composite pores. The soil parameters required to estimate a radon diffusion coefficient are thus the moisture, the packing density, and the pore size distribution.

The following sections contain a mathematical description of radon diffusion in both homogeneous and two-phase media, the development of a complex pore model, and the application of the model to real soils. The model development section includes descriptive soil parameters, open and water-blocked pore models, a description of moisture distribution in the various pores, a random pore combination model, and a description of pore blockage as saturation is approached. The application section discusses determination of pore size distributions, the computer code used to calculate the diffusion coefficients, the results of varying parameters and comparisons with measured diffusion coefficients.

MATHEMATICAL DESCRIPTION OF RADON DIFFUSION

Radon diffusion in soils occurs in both the air-filled and water-filled regions of pore space. Similar diffusion equations can be used to describe each phase, which can be combined with appropriate distribution coefficients to describe the dual system. The diffusion equations are described below for a single phase system, and are then combined in describing the water-air mixture for soil pores.

Diffusion in Homogeneous Media

In diffusion theory the diffusion coefficient is defined by Fick's law, which relates the radon flux to the radon concentration gradient. For one-dimensional diffusion this is

$$J = -D_e(\partial C/\partial x) \ , \tag{1}$$

where

J = radon flux from the bulk porous material (pCi/cm^{2}s)

C = radon concentration in the total pore space of the material (pCi/cm^3)

D_e = effective bulk diffusion coefficient of the porous material (cm^2/s)

Lack of uniformity exists in the literature with respect to the appropriate diffusion coefficient to use in predicting radon migration. The confusion arises from differing definitions of C and J as well as from the nature of common radon measurement methods. Radon concentrations sampled from within the soil are interstitial quantities, and hence the concentrations are measured per unit volume of pore. Conversely, the radon flux is measured across the total or bulk soil surface and not just across the exposed interstices(11). Figure 1 illustrates these bulk and interstitial quantities for a schematic cross section of a soil.

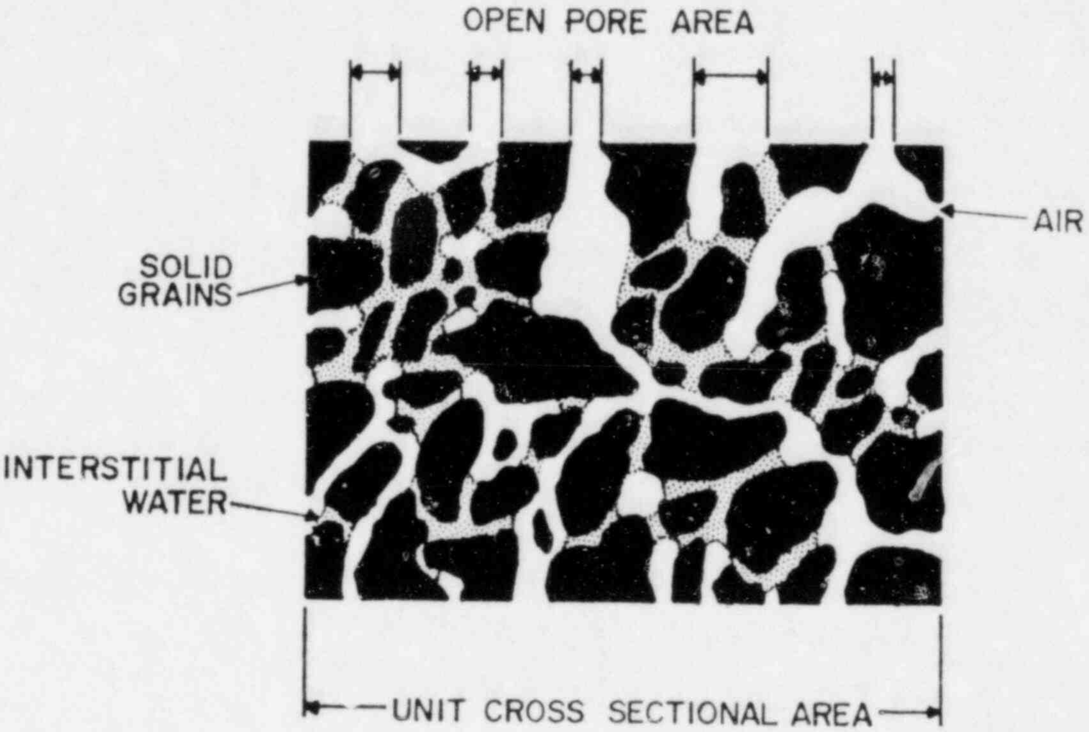

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FIGURE 1. REFERENCE VOLUME OF UNSATURATED POROUS MATERIAL

Use of D_e to relate the radon concentration gradient in the pore space to the bulk flux, J, accounts for the restricted nature of the channels in which diffusion occurs in the porous medium. In comparison to diffusion in a continuous or homogeneous medium, the two main factors which reduce the radon flux from a porous material are the reduction in cross-sectional area for diffusion, and secondly, the lengthening of the pathway for diffusion due to the circuitous nature of the pores.(14)

It is often helpful to refer to the radon diffusion coefficient of the interstitial or pore space when describing radon diffusion through a porous earthen material. The diffusion coefficient of the pore fluid can be shown to be related to the effective bulk diffusion coefficient by

$$D = D_e/P \ , \tag{2}$$

where D = diffusion coefficient of the pore fluid (cm^2/s)

P = total porosity of the porous medium.

The pore diffusion coefficient is defined from Fick's law as

$$j = -D\ (\partial C/\partial x) \tag{3}$$

where j = radon flux in the pore space (pCi/cm^{2}s).

Although j is not a measurable quantity, it can be visualized from Figure 1 as the radon flux per unit open-pore area. It is related to the measurable flux per unit area of the bulk soil by

$$J = Pj. \tag{4}$$

In order to clearly visualize the foregoing definitions of diffusion coefficients and also those which follow, Figure 2 illustrates these definitions. As illustrated, the measurable pore diffusion coefficient, D, may be a complicated function of various configurations such as air-filled channels, air and water regions, and water-blocked channels. However, in the respective limits of complete dryness or complete saturation, D may be related to the bulk diffusion coefficients of air or water by

$$D_a = \tau D_A \tag{5}$$

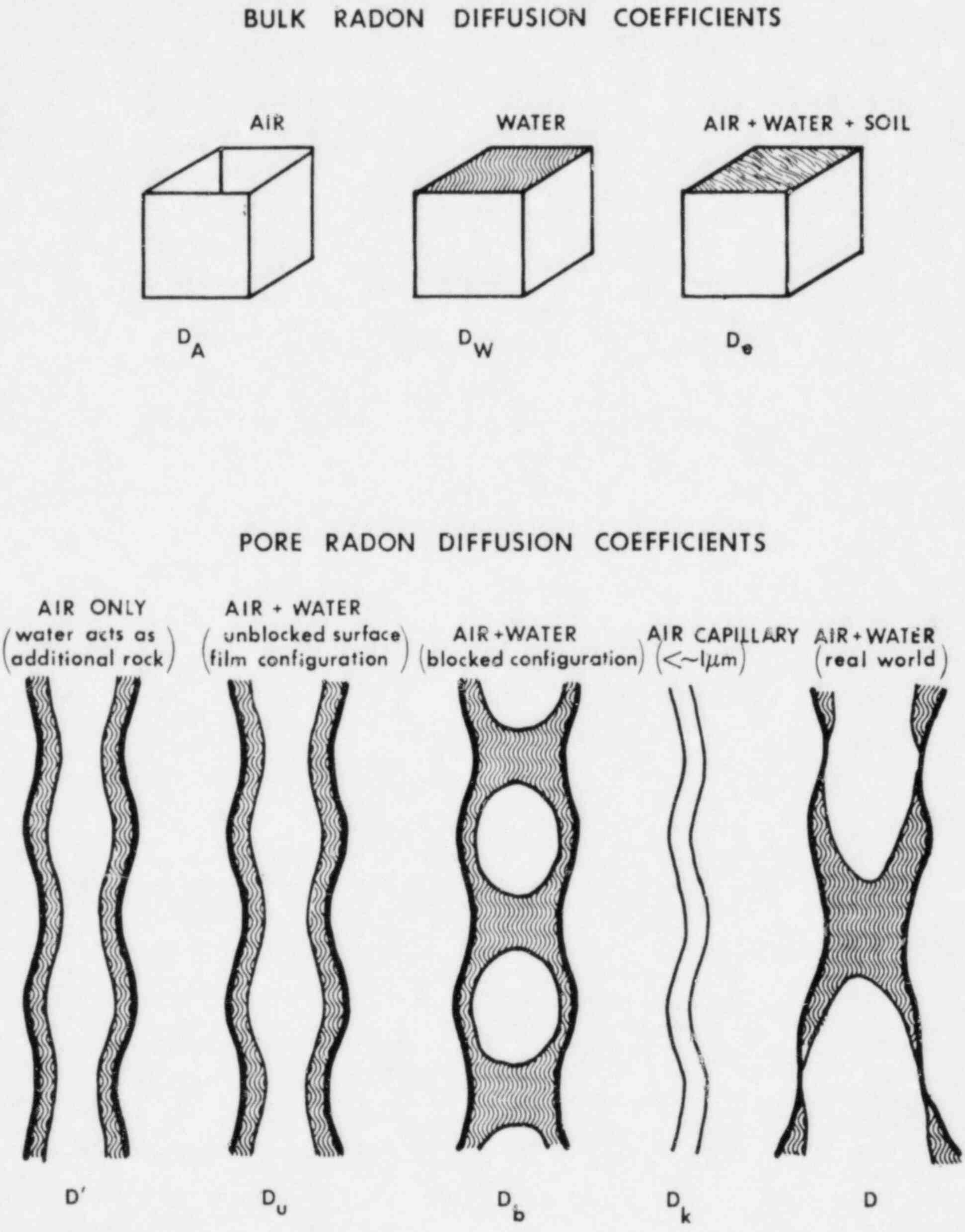

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FIGURE 2. ILLUSTRATION OF DEFINITIONS OF RADON DIFFUSION COEFFICIENTS

and $$D_w = \tau D_W \qquad (6)$$

where D_a = pore radon diffusion coefficient in a dry, air-filled porous medium (cm^2/s)

D_w = pore radon diffusion coefficient in a water-saturated porous medium (cm^2/s)

τ = tortuosity of the porous medium = $(L/L_e)^2$, L = bulk medium thickness, L_e = diffusion path length. A typical value of τ is 0.66, as obtained for uniform close-packed spheres.

D_A = radon diffusion coefficient of pure air(31) (0.11 cm^2/s)

D_W = radon diffusion coefficient of pure water(3) (10^{-5} cm^2/s)

Reference 14 contains an excellent review of the nomenclature and definitions of the diffusion coefficients reported in the literature prior to 1976, which are consistent with the present definitions. However, a recent pertinent review article(19) gives a relationship different than that in Equation 2.

While most authors(3,12,14,22,23,32) describe radon diffusion and the associated Fick's law relationship using the radon concentration in the pore space, C, some authors(11,33) use the radon concentration per unit volume of bulk porous material. These two are simply related by

$$C_e = P\,C \qquad (7)$$

where C_e = radon concentration in the bulk porous soil medium (pCi/cm^3).

Substitution of Equations 2 and 7 into Equation 1 yields the following interesting result:

$$J = -D(\partial C_e/\partial x) \ . \tag{8}$$

Therefore, when the gradient of C_e is related to the bulk radon flux, the diffusion coefficient of the fluid in the pore space is obtained.

An explicit consideration of radon migration only through the air-filled component of the pore space(29) utilized the following Fickian relation:

$$J = -D_e'(\partial C_a/\partial x) \ , \tag{9}$$

Where C_a = radon concentration in the gas-filled pore space.

At dryness or low moistures, C_a is similar to C, so that D_e' is similar to D_e. At high moistures, the assumption that radon only diffuses in the air-filled pore space becomes questionable and it is more difficult to relate D_e' and D_e. The subject and implications of radon diffusion in only the air-filled pore space are still under investigation.

When Fick's law relationships are incorporated into the radon balance equation, the form of the diffusion equation containing D_e will also contain an explicit reference to the porosity of the bulk material, while in the balance equation containing D, explicit reference to the porosity only occurs in the source term for those materials containing sources. Because of this feature, the remainder of the present discussion will utilize only pore diffusion coefficients, D, not D_e. Subscripts that will be associated with the pore diffusion coefficients refer to specific values or specific models for D, as illustrated in Figure 2.

Diffusion in Two-Phase Media

Radon diffusion through a two-phase medium in the pore spaces of earthen materials involves several complex processes. There is radon diffusion through the air in the presence of a water layer. There is radon diffusion through the water in water-blocked pore regions, and there is radon absorption in the water, as well as other mechanisms. The general objective of the present effort is to characterizize these mechanisms which occur on a microscopic scale by a single macroscopic diffusion coefficient which can be used to describe the radon diffusion as if occurring through a simple homogeneous medium characteristic of the porous material.

Before proceeding with the differential equation describing diffusion, it is instructive to discuss the nature of the source term, Q. In general it is given by:

$$Q = R\,E\,\rho\,\lambda/P\ , \tag{10}$$

where Q = volumetric radon source strength (pCi/cm^{3}s)

R = specific radium activity (pCi/g dry mass)

E = emanation coefficient (pCi released into pores/total pCi produced)

ρ = dry density of material (g/cm^3)

λ = decay constant for radon (2.1×10^{-6} s^{-1})

When considering the emanation of radon into the pore space, the question arises of whether the emanation occurs into the air or water-filled component of the space. Emanation has been measured and modeled for both rock-air and rock-water interfaces, and also described for intermediate, partially-saturated systems(34). Reference 34 also contains a comprehensive bibliography on the effects of moisture on radon emanation and diffusion. In general the total E increases linearly with moisture from a value of E_a until a plateau moisture m^* is reached, at which point E remains a constant E_w until the material is saturated. The air and water components of E can be described by the following relationships:

$$E = E_{air} + E_{water} , \tag{11}$$

where E_{air} is the fraction of the radon generation that occurs in the air component of the pore space and E_{water} is the fraction of the radon that occurs in the water component of the pore space. The E_{air} and E_{water} terms are given by:

$$E_{water} = \begin{cases} \frac{m}{m^*} E_w & \text{for } m < m^* \\ E_w & \text{for } m > m^* \end{cases} \tag{12}$$

$$E_{air} = \begin{cases} E_a(1 - m/m^*) & \text{for } m < m^* \\ 0 & \text{for } m > m^* \end{cases} \tag{13}$$

where m = volume fraction of moisture saturation

E_a = emanation coefficient at dryness

E_w = emanation coefficient at saturation

m^* = minimum moisture on plateau of emanation/moisture curve

The constants m^*, E_a and E_w are constants from the emanation model.

The one-dimensional, steady-state diffusion of radon through a moist, unsaturated earthen material can be described as the combined diffusion in the gas- and liquid-filled pore volumes of the material. The gas-phase diffusion can be described by

$$D_a \frac{d^2C_a}{dx^2} - \lambda C_a + \frac{R\rho\lambda E_{air}}{P_{air}} + T_{wa} = 0 \qquad (14a)$$

where D_a = the diffusion coefficient in the air-filled pore space (cm^2/s)

C_a = the concentration of radon in the air (pCi/cm^3)

T_{wa} = radon transfer rate from the water to the air (pCi/cm^{3}s)

P_{air} = air-filled porosity, = P(1-m)

The corresponding equation for diffusion in the liquid-filled pore region is:

$$D_w \frac{d^2D_w}{dx^2} - \lambda C_w + \frac{R\rho\lambda E_{water}}{P_{water}} - T_{wa} = 0 \qquad (14b)$$

where D_w = the diffusion coefficient in the water filled pore spaces (cm^2/s)

C_w = the concentration of radon in the water-filled space (pCi/cm^3)

P_w = water-filled porosity = Pm.

It is assumed that D_a and D_w are spatially independent. The radon concentration in the total pore space is the volume-weighted average concentrations in the air and water components. It is given from the previous definitions of C_a and C_w by

$$C = C_a\ (1-m) + C_w\ m \qquad (15)$$

Equation (15) provides the basis for describing radon diffusion in the combined air and water phases of the pore space. If Eq (14a) is multiplied by (1-m) and Eq (14b) is multiplied by m and then the two equations are summed, the result is

$$D_a \frac{d^2C_a\ (1-m)}{dx^2} + D_w \frac{d^2C_w m}{dx^2} - \lambda C_a(1-m) - \lambda C_w m + \frac{R\rho\lambda}{P}\ (E_{air} + E_{water}) = 0 \qquad (16)$$

In forming the combined equation it is assumed that m is spatially independent. It is also noted that the radon transfer rate cancels, because a transfer from water to air in the pore space is no longer a source or a sink term. The dispersion term is used to define the combined D.

$$DC = D_a C_a (1-m) + D_w C_w M.$$

The resulting total pore diffusion equation, assuming intimate mixing of the air and water, but no total passage blockage by water, is given by

$$D \frac{d^2C}{dx^2} - \lambda C + R\rho\lambda E/P = 0 \qquad (17)$$

The diffusion coefficient D, which represents the combined "real-world" diffusion coefficient of the unsaturated pore, depends on the relative volumes of air and water in the pore space, and on their physical configurations. In principle, D is a function of m, D_A and D_W; however, the functional relationship is complicated and has not been defined. It has been approximated with a model using combinations of unblocked and water-blocked pores, however, that is described in the following section.

PORE MODEL DEVELOPMENT

In order to mathematically describe the diffusion of radon through a porous medium, the medium must be characterized with respect to (a) the pore fluid and (b) the pore structure. This section provides the basis for characterizing earthen materials in both of these respects. The characterization makes use of configurational models, but is based directly on measured physical parameters of the earthen materials.

Several soil parameters are potentially related to radon diffusion coefficients and could potentially be used as a basis for a diffusion model. These include moisture, bulk density, total porosity, air-filled porosity, pore size distribution, grain size distribution, air permeability, hydraulic conductivity, internal surface area, and perhaps others. Since many of these parameters are interrelated, only those providing a unique contribution to the material description were utilized. The following model development is based only on the moisture content and the pore size distribution of the earthen material. Other parameters needed in the calculations can be derived from these. For example, the total porosity is obtained by integrating the pore size distribution; and the bulk density (ρ) can be calculated from the total porosity (P) by assuming a typical specific gravity (SG) for western cover materials of 2.7 g/cm^3,

$$\rho = SG\ (1-P) \tag{18}$$

Pore Fluid Configuration

The pore fluid is composed of water and air, whose relative volumes are defined by the moisture content and total porosity of the soil. The configuration of the water-air mixture in the pores is extremely important in its impact on the diffusion coefficient of the pores. Two extreme water-air configurations are illustrated in Figure 2, as represented by D_u and D_b. The D_u configuration assumes the water to occur as a film covering the surface of the pore, but not blocking its central air-filled channel. This configuration is probably a good approximation of the major pores in a soil. The small intragranular pores become saturated first, leaving the larger intergranular pores with moist or water-covered surfaces. The radon diffusion coefficient for the D_u configuration is given by

$$D_u = \frac{D_a}{1 + \frac{mk}{1-m}} + \frac{D_w}{1 + \frac{1-m}{mk}} , \tag{19}$$

where k = radon distribution coefficient for water/air
(k = 0.26 at 20°C).

The u subscript refers to the fact that the total passage is unobstructed by water blockages as illustrated in Figure 2.

Equation 19 can also be shown to be valid for a hypothetical, homogeneous mixture of air and water in a pore, which is mathematically equivalent to the surface-film configuration D_u in Figure 2. Although the air-water mixture in a pore is not likely to occur in a continuous, homogeneous configuration, the simplicity of its description makes it useful in deriving Equation 19 for the D_u configuration.

Although many intergranular pores may be represented by the D_u configuration, pore blockage by water, as illustrated in Figure 1, is also significant. An extreme representation of a water blocked pore is illustrated by the D_b configuration in Figure 2. If all of the water were to occur as intermittent pore blockages, the diffusion coefficient can be estimated by solving the resulting multilayer system. For this case, $C_a = C$ in the air region and $C_w = C$ in the water region. Solution of the equations yields an expression of the form of Equation 17, in which the diffusion coefficient, represented by D_b, is given by

$$D_b = \frac{D_w}{[m+(1-m)(D_w/D_a)^{\frac{1}{2}}]^2} \tag{20}$$

Although a soil system is not comprised entirely of either unblocked or blocked pores, these two extremes define the probable maximum and minimum diffusion coefficients for a soil system. Figure 3 illustrates the relative magnitudes of the unblocked (D_u) and blocked (D_b) pore diffusion coefficients in comparison with measured diffusion coefficients reported for a variety of soils. As this comparison suggests, soil pores can be described by a combination of unblocked and blocked pores. The value of D for a particular unconsolidated material depends upon the physical configuration of the pore spaces as well as other factors. If the configuration is such that significant pore blockage occurs with relatively little moisture, then the D value will tend to be closer to D_b. Conversely, if the soil can absorb a relatively large quantity of water before pore blockage occurs, then the value of D will be closer to D_u.

In general unconsolidated earthen materials have pore spaces randomly distributed in volume and location; thus, a random, statistical analysis of pore configuration is suggested and is described in the next section. Certain geological samples may be characterized by a quasi-ordered pore configuration and their analysis must be modified accordingly.

The value of D_a used in Equations 19 and 20 includes the effect of the air-filled pore diameter. As the pore diameter decreases, Knudsen diffusion

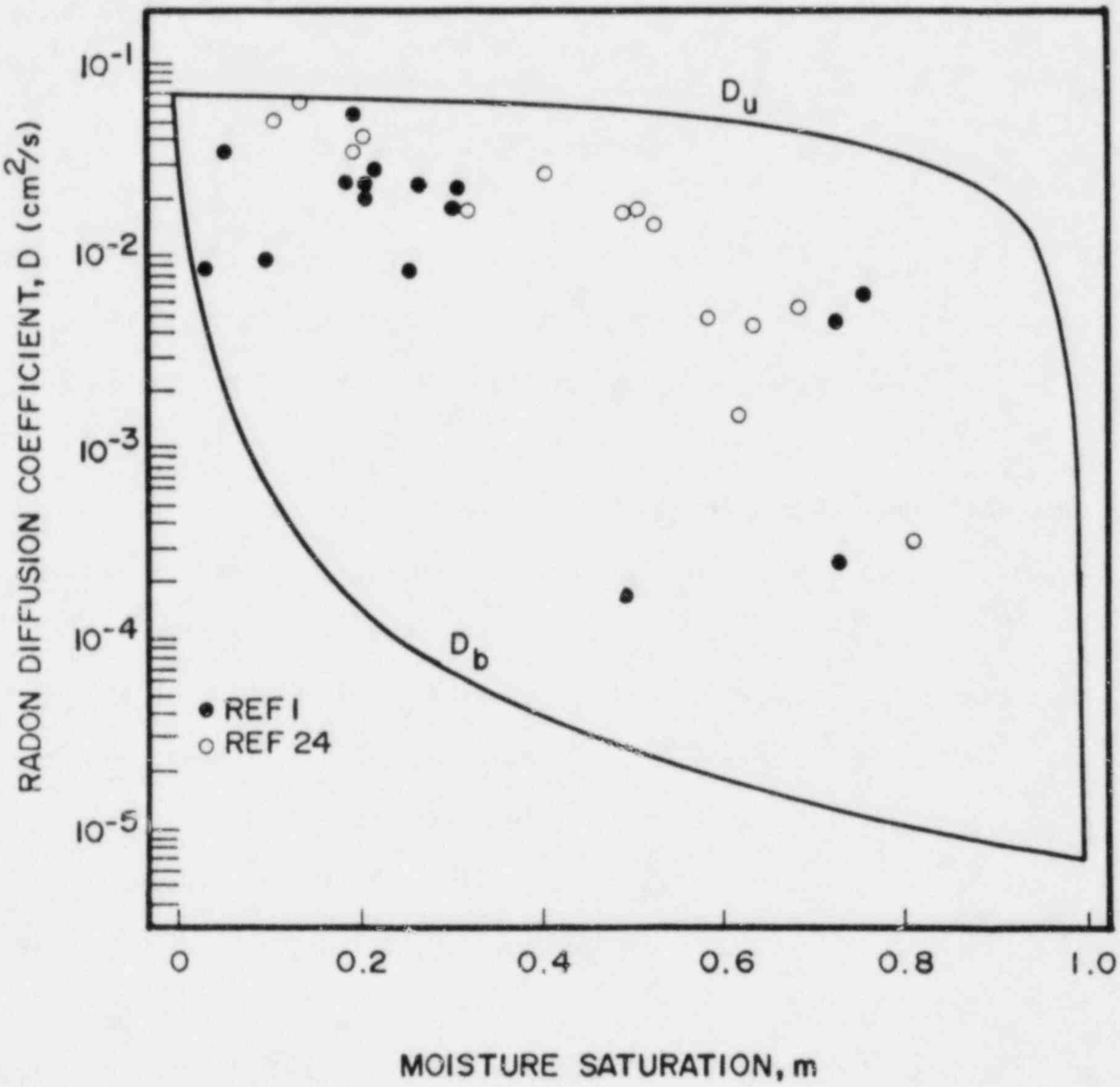

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FIGURE 3. COMPARISON OF DIFFUSION COEFFICIENTS FOR WATER-BLOCKED PORES, D_b, AND UNBLOCKED PORES, D_u, WITH MEASURED DIFFUSION COEFFICIENTS.

becomes dominant.(35) The expression for D_a which includes the Knudsen and transition realm is:(36)

$$\frac{1}{D_a} = \frac{1}{\tau D_k} + \frac{1}{\tau D_o} \tag{21}$$

where D_k = Knudsen diffusion coefficient = 1.12 r

r = radius of air-filled space in pore (μm)

In addition to describing the physical configuration of the water in pore spaces, the sequence in which pores of various sizes absorb water must also be defined. It is recognized that capillary effects cause small pores to absorb water before large pores. However, since large pores are usually intergranular and are lined by smaller pores, the large pores may also contain an effective film of water prior to the saturation of the smaller pores.

The pore filling sequence chosen for the present model was therefore defined by considering a water film of uniform thickness to be present in all pores. The film thus caused a greater degree of saturation in small pores than in large ones. When the water film thickness equalled or exceeded the radius of a measured pore size increment, that increment was considered saturated. Additional water from thicker films was added only to the pore size increments which were still unsaturated. The volumetric saturation of a soil system was therefore calculated from a given water film thickness and pore size distribution as

$$m = \frac{\sum_i v_i \left[1-(r_i-w)^2/r_i^2\right]}{\sum_i v_i} , \tag{22}$$

where v_i = incremental volume of pores of radius r_i(the measured pore size distribution)

w = the assumed water film thickness (μm)

Ideally, Equation 22 should express w as a function of m. Since the equation cannot be explicitly solved in this form, however, the value for w at a given m is determined by successive approximation.

An additional refinement was included in the above pore-filling sequence. This was to account for the fact that an extremely small air-filled opening is not stable in a water-filled annulus. Instead, the water will completely block or saturate a pore segment when w is almost as large as r_i. The minimum radius of an air-filled opening in a pore was estimated from the pressure-curvature relationships which characterize capillary effects. By combining the bubble pressure-radius relationship(37) with the capillary suction expression for a pore,(38) the following relationship was derived:

$$r_c = (0.7 + 1/r_i)^{-1} , \qquad (23)$$

where r_c = the minimum radius of curvature of an air-filled region of a pore (μm)

r_i = the pore radius (μm)

Using the above descriptions of water configuration, a radon diffusion coefficient can be calculated for any pore size increment at any moisture content using Equation 19 and Equation 22. Although Equation 19 only considers unblocked pores (D_u, Figure 2), the effect of pore blockage by water is considered by combining saturated pore segments in series with unsaturated segments.

The method of combining the diffusion coefficients for each pore size increment is presented in the following section.

Pore Structure

The manner in which the pore structure of the earthen material is modelled from the measured pore size distribution is critical in obtaining the proper diffusion coefficient-moisture relationship. For example, a model which assumes the distribution of pores to consist of parallel cylindrical tubes of various diameters has been reported.(32,36) However, the radon diffusion coefficients predicted by such a model resemble the D_u curve in Figure 3, overestimating the diffusion coefficients for most soils due to the dominance of the large pores. Converging or diverging composite pores which incorporate all pore sizes in series have also been proposed.(36) However, the early blockage of the small pores as moisture is added to the composite series retards diffusion in the entire pore, resulting in low diffusion coefficients, similar to the D_b line in Figure 3.

A completely random pore combination model was used to describe the earthen materials in this work. It was recognized that all sizes of pores may be interconnected to each other, and that some diffusion paths may resemble straight cylinders while others may resemble narrow-necked chambers which would suffer from intermittent water blockage. The measured pore size distribution was therefore combined statistically to produce all possible combinations of single and composite pores. A simplified example of the model pore combinations is illustrated for a three-component pore size distribution in Figure 4. In cases of composite pores, the relative heights of each segment were determined from the relative total lengths of the given pore components as calculated from the pore size distribution. The total length of each pore size increment was calculated as

$$h_i = \frac{v_i}{\pi r_i^2} \quad . \tag{24}$$

The diffusion coefficients determined for each pore or pore segment were combined in all single and composite pore combinations, and the resulting pores, as illustrated in Figure 4, were considered to exist in parallel in the actual soil. As illustrated, composite pores provided more possible combinations than single pores, and were weighted with a greater total volume. Typically, nine pore size increments were incorporated in calculating a radon diffusion coefficient.

The diffusion coefficients for each of the composite pores (D_{cp}) were calculated from the diffusion coefficients of their segments using the weighting function

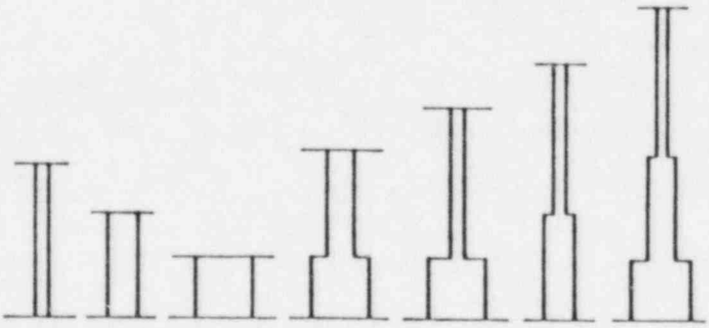

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FIGURE 4. ILLUSTRATION OF PORE COMBINATIONS FOR A THREE-COMPONENT PORE SIZE DISTRIBUTION

$$D_{cp} = [\sum_i f_i/(D_{ui})^{\frac{1}{2}}]^{-2} \tag{25}$$

where f_i = the fractional height of the pore segment in the composite pore

D_{ui} = The individual segment diffusion coefficients calculated from Equation 19

The resulting diffusion coefficients for all possible single and composite pores were then weighted according to the volume or combined volumes of the given pore sizes to estimate the average diffusion coefficient for the pores of the earthen material:

$$D = \frac{\sum_j (D_{cpj} \sum_i^k v_i)}{\sum_j \sum_i^k v_i} , \tag{26}$$

where D = the effective radon diffusion coefficient of the material, as defined by Equation 2 and Equation 8.

j = subscript referring to each single or composite pore.

i = subscript referring to each incremental pore size.

k = number of size increments in composite pore j.

PORE MODEL APPLICATION

The above pore structure and moisture models were tested and evaluated using a computer code to calculate radon diffusion coefficients from the pore size distribution and moisture contents of various soils. This section describes the determination of the soil parameters required for these calculations and the computer code used for the calculations, and compares the results with published correlations and measured diffusion coefficients.

Determination of Soil Parameters

Soil moisture content is a widely measured parameter which has been used previously to predict diffusion coefficients.(22,29) It is accurately determined by gravimetric methods and is usually expressed as a dry weight percentage. The volumetric moisture saturation, m, is determined from the dry weight percentage (M) as

$$m = \frac{M\rho}{P} \tag{27}$$

The pore size distribution depends on the particle size and compaction of the earthen material, and can be estimated from these parameters or measured by mercury intrusion porosimetry or, for small pores, by gas adsorption characteristics.(39) The pore size distribution is often more conveniently determined from water retention characteristic curves(38,40) which are measured to define the hydrologic properties of soil systems. In making these estimates the following relationship is used to relate pore radius to capillary pressure:(38

$$r_i = -\frac{1480}{\Psi} \tag{28}$$

where r_i = pore radius in μm

Ψ = soil capillary pressure in cm of water

Procedures for differentiating the water retention curve are similar to those used with mercury intrusion porosimetry curves.(39)

The direct calculation of pore size distributions from grain size and density data is theoretically attractive because of the common availability of sieve analyses and compaction data. Attempts have been made to relate grain size distributions to a pore size distribution index(41) in estimating hydraulic conductivities of soils. A more recent report has given a more direct correlation of pore size and grain size distribution parameters.(42) It is expected that such applications of direct pore size distribution calculations may yield data of sufficient accuracy for diffusion coefficient calculations, particularly in the absence of more reliable water retention curve data.

The tortuosity is primarily a function of the soil porosity and of the width of its pore size distribution. Although natural soils are often well represented by the constant value $\tau = 0.66$, a more detailed calculation is used in the present model to accommodate the wide range of parameters being considered. The determination of τ is partially based on considerations of uniform packed spheres, from which the following relationship with porosity was derived:

$$\tau = \left\{1 + \left[\frac{3(1-P)}{2\pi}\right]^{2/3}\right\}^{-1} \tag{29}$$

This equation accounts for the less-tortuous diffusion pathway which occurs when uniform spheres are less-densely packed, and reaches the most tortuous value, $\tau = .66$ for the close-packed geometry when $P = 0.26$.

When spheres of different sizes are mixed, small spheres may fill voids between larger spheres. In close-packed geometry, a binary mixture of very large and very small spheres would have the tortuosity of each size superimposed, resulting in a combined tortuosity of $\tau = (0.66)^2$. If three very different sizes were mixed, the exponent could become 3, or in general, equal to the width of the grain size distribution normalized to the ratio of inscribed sphere radii. This ratio is 11.3 for close-packed spheres, and using the grain-size to pore-size model of Arya and Paris,$^{(42)}$ becomes 44 if the width of the pore-size distribution is used instead of that for the grain size distribution. The resulting tortuosity expression used in the following diffusion model calculations is

$$\tau = \left\{1 + \left[\frac{3(1-P)}{2\pi}\right]^{2/3}\right\}^{-(1+2GSD/44)} \tag{30}$$

where GSD = geometric standard deviation of the pore-size distribution

Calculations

Radon diffusion coefficients were calculated by a simple FORTRAN computer code which used a nine-increment pore size distribution and the soil density and porosity as input. The pore size distribution was first normalized to the total porosity of the soil, after which total pore lengths were computed for each pore size increment using Equation 24. A loop was then entered to calculate the radon diffusion coefficients at each of 26 different moisture contents. The moistures varied for each sample, and were determined from a corresponding array of water film thicknesses ranging from 0.001 μm to 2000 μm. Values of m were calculated from Equation 22.

At each given moisture content, a diffusion coefficient was calculated for each pore size increment using Equation 19. When the air-filled volume of the pore had a small (<10 μm) radius, the resulting diffusion coefficient was reduced accordingly with Equation 21 to account for Knudsen and transitional diffusion. If the air-filled radius of the pore was smaller than the critical radius of curvature for the given pore size (Equation 23), the pore increment was considered saturated, and assigned a diffusion coefficient equal to D_w.

When the diffusion coefficient was defined for each pore size increment, the diffusion coefficients for all possible single and composite pores were calculated using Equation 25, which is a height-weighted equation for serial pore combinations. The resulting 511 diffusion coefficients were then combined by the volume-weighting function, Equation 26, for parallel diffusion through single and composite pores. This entire calculation sequence was then repeated for a different water film thickness and continued until 26 diffusion coefficients and moisture contents had been calculated.

Results

Radon diffusion coefficients were calculated from the random pore combination model for several arbitrary pore size distributions to evaluate the sensitivity of the diffusion coefficient-moisture relationships to pore sizes. Calculated diffusion coefficients were also compared to measured diffusion coefficients for soils whose pore sizes were calculated from moisture drainage curves. The calculated diffusion coefficients were also compared to results predicted by two published correlations with porosity and moisture. The following paragraphs summarize these results.

Sensitivity to Particle Size Parameters

The sensitivity of calculated diffusion coefficients to median particle size and to the geometric standard deviation (GSD) of the pore size distribution was examined. Nine pore size distributions were calculated to simulate a wide range of possible soil materials. These are summarized in Table I. As indicated, the median pore diameter was held constant in six of the distributions, and the geometric standard deviation of the pore size distribution was held constant among four other distributions. The diffusion coefficients calculated from these distributions assuming a total porosity of 0.4 are plotted in Figures 5 and 6.

As indicated in Figure 5, radon diffusion coefficients decreased with decreasing median pore size. The largest decreases were observed in the dry range, where about an order of magnitude difference in diffusion coefficient was attributed to the differences in median pore size. This difference is likely due to the dominance of Knudsen diffusion in the small pore size ranges. Typical earthen cover materials are probably best approximated by the 1-100 μm median pore size curves. The geometric standard deviation of the pore size distribution was fixed at 5.0 for these curves.

Figure 6 illustrates a similar comparison in which the geometric standard deviation of the pore size distribution was allowed to vary. As indicated, diffusion coefficients decreased systematically with increasing GSD. Typical soil materials are generally in the 2-10 range of GSD, although unusually well-sorted materials may exhibit even lower values. As illustrated, the wide variation in GSD of the pore sizes caused an approximate two order-of-magnitude variation in D.

Comparison with Measured Data

The random pore combination model was evaluated with eight sets of pore size distribution data obtained from water retention curves as described in

Table I

Pore Size Distributions Used for Diffusion Coefficient Sensitivity Tests

v_i (10^5cm^3/g)	r_i (μm)								
49	10	1.1	0.058	0.0063	0.00019	0.0000056	0.00058	0.0058	0.58
422	10	1.9	0.21	0.040	0.0029	0.00020	0.0021	0.021	2.1
2087	10	3.3	0.76	0.25	0.043	0.0075	0.0076	0.076	7.6
5452	10	5.7	2.8	1.6	0.66	0.27	0.028	0.28	28
7509	10	10	10	10	10	10	0.10	1.0	100
5452	10	17	36	63	152	366	0.36	3.6	360
2087	10	30	131	398	2,310	13,400	1.31	13.1	1,310
422	10	53	480	2,510	35,100	490,000	4.8	48	4,800
49	10	92	1,720	15,800	533,000	17,900,000	17.2	172	17,200
Median r	10	10	10	10	10	10	0.1	1	100
GSD	1	2	5	10	30	90	5	5	5

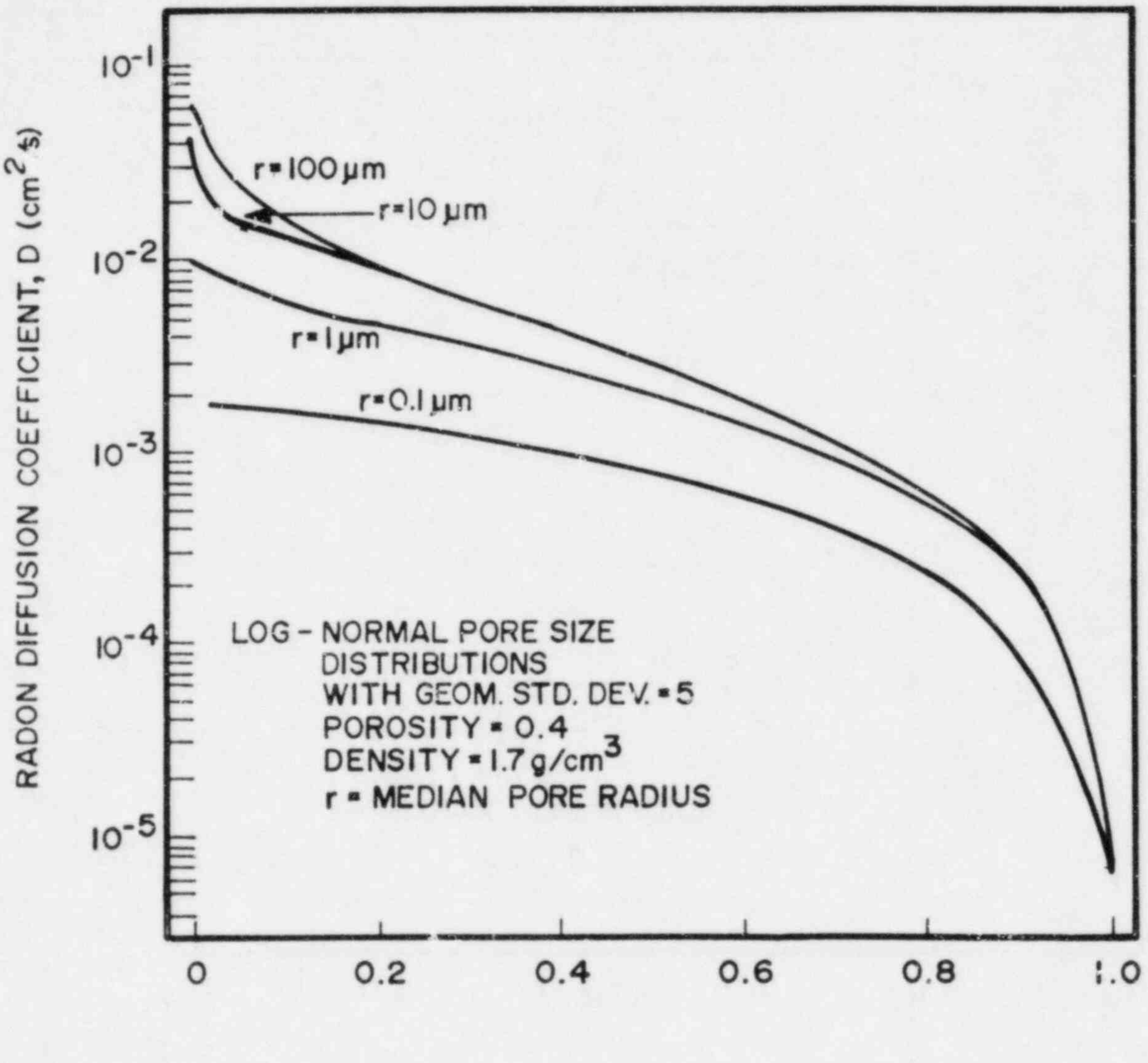

FIGURE 5. COMPARISON OF DIFFUSION COEFFICIENT MOISTURE CURVES FOR VARIOUS MEDIAN PORE SIZES.

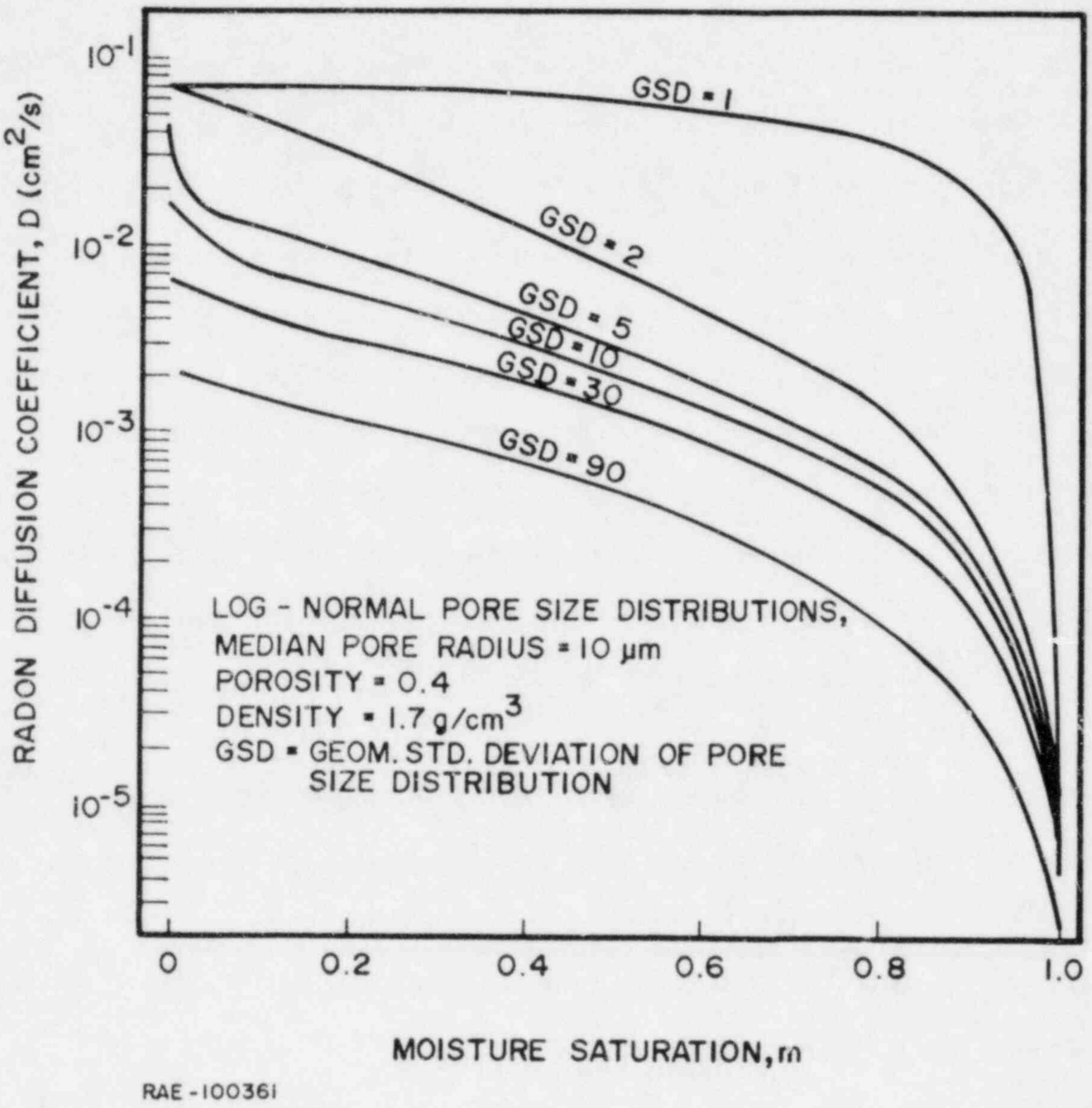

FIGURE 6. COMPARISON OF DIFFUSION COEFFICIENT - MOISTURE CURVES FOR VARYING WIDTHS OF PORE SIZE DISTRIBUTIONS.

Reference 38. The water retention curves and measured diffusion coefficients, for comparison, were taken primarily from Reference 24 with some additional measurements made as part of the present work. The measured diffusion coefficients are plotted in Figure 7 as a function of the moisture content of the soils. As indicated, nearly all of the measured diffusion coefficients fall within the shaded region which defines the range of calculated diffusion coefficients for the eight soils. A particular diffusion coefficient curve for soil VC is also plotted, for comparison with the ten measured diffusion coefficients for this soil. By comparing the measured data points with the calculated curves for soil VC and the other soils, the relative RMS error between the measured and predicted diffusion coefficients was estimated to be about 50 percent excluding the two open-circle points. The open circles represent measured points in question which were determined to be excessively low in the original report.(24)

If the soils used to evaluate the model in Figure 7 are assumed to have pore size distributions typical of other earthen materials under similar test conditions, a comparison can be made with the previously reported diffusion coefficients plotted in Figure 3. Although a somewhat greater scatter in the data is observed, the data are generally within the same range. The lack of pore size information on the earlier data points precludes further interpretation and comparison with model predictions.

Comparison with Published Correlations

Two simple correlations have been reported for estimating radon diffusion coefficients from moisture or air-filled porosity. These are based on the fact that for western soils with low moisture contents, most of the measured values

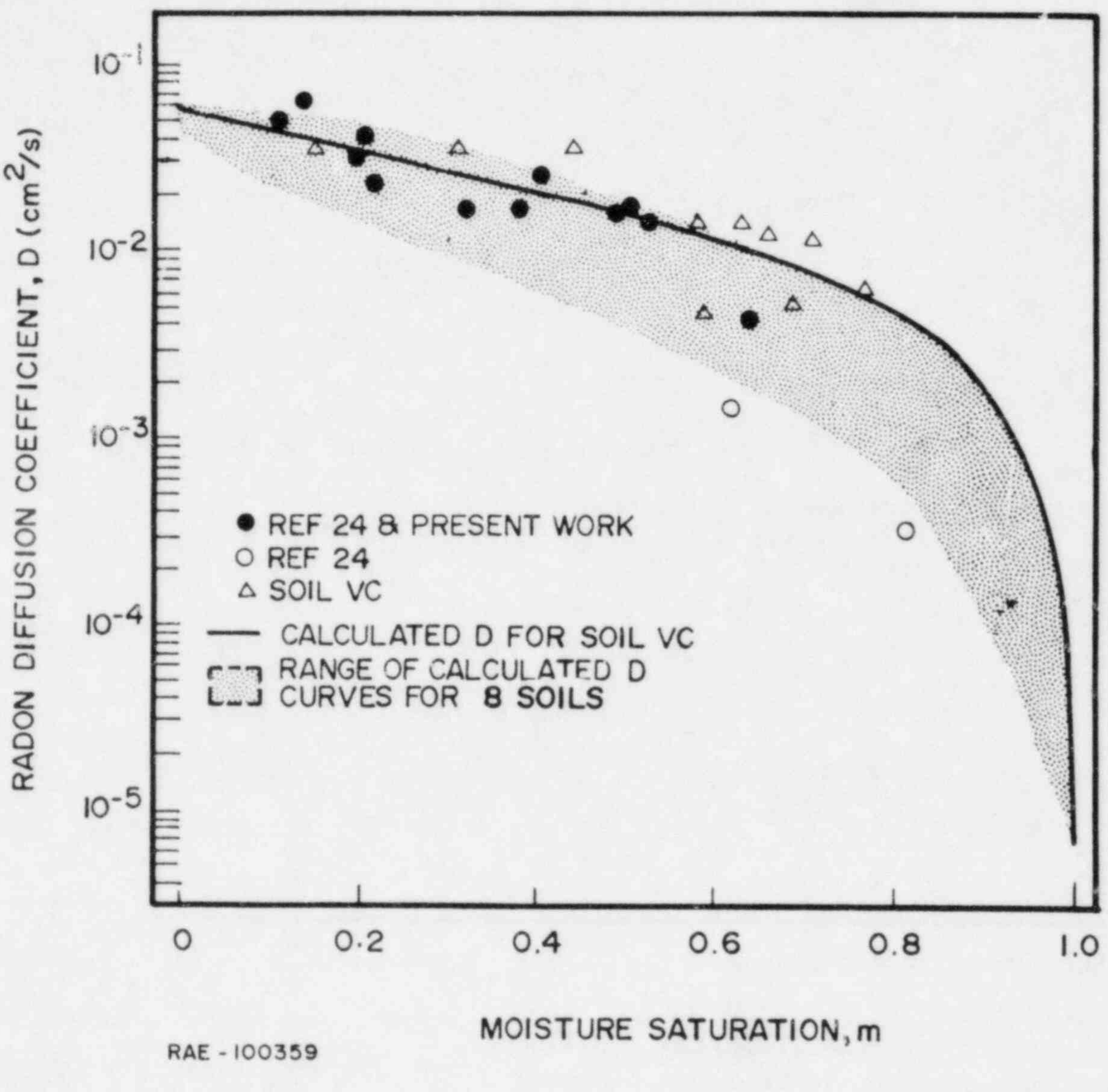

FIGURE 7. COMPARISON OF MEASURED RADON DIFFUSION COEFFICIENTS WITH CALCULATED VALUES.

of D at a given m are within a factor of 3 of each other.

It was reported in Reference 22 that the diffusion coefficients for a wide variety of western soils could be approximated by the following simple correlation involving moisture and the diffusion coefficient of radon in air:

$$D = D_A \exp(-0.261M') \quad , \qquad (31)$$

where M' = weight-percent of moisture in the porous material (i.e., grams water per gram wet sample)

The same data were used in Reference 29 to obtain a correlation similar in form to other correlations of gaseous diffusion through porous materials.(30) The correlation based upon the air-filled porosity, p_a, is:

$$\frac{D_e}{P_a} = D' \; 0.74 \; D_A \; p_a^{1.16} + \tau D_W \frac{m}{(1-m)} \qquad (32)$$

where $\tau = 0.66$

The correlations represented by Equations 31 and 32 are given in Figure 8 assuming a fixed total porosity of 0.4 and, for applying Equation 31, a bulk density of 1.7 g/cm^3. These correlations have been shown to agree with the data given in Figure 3 and lie well within the bounds defined by the D_u and D_b curves, which are also shown in Figure 8. It should be noted that the A and B curves will be shifted for different values of P and ρ, and that the plotted curves are merely typical of these soil parameters.

Since the correlations were estimated from a variety of measured diffusion coefficients without regard to pore size distributions, specific comparisons

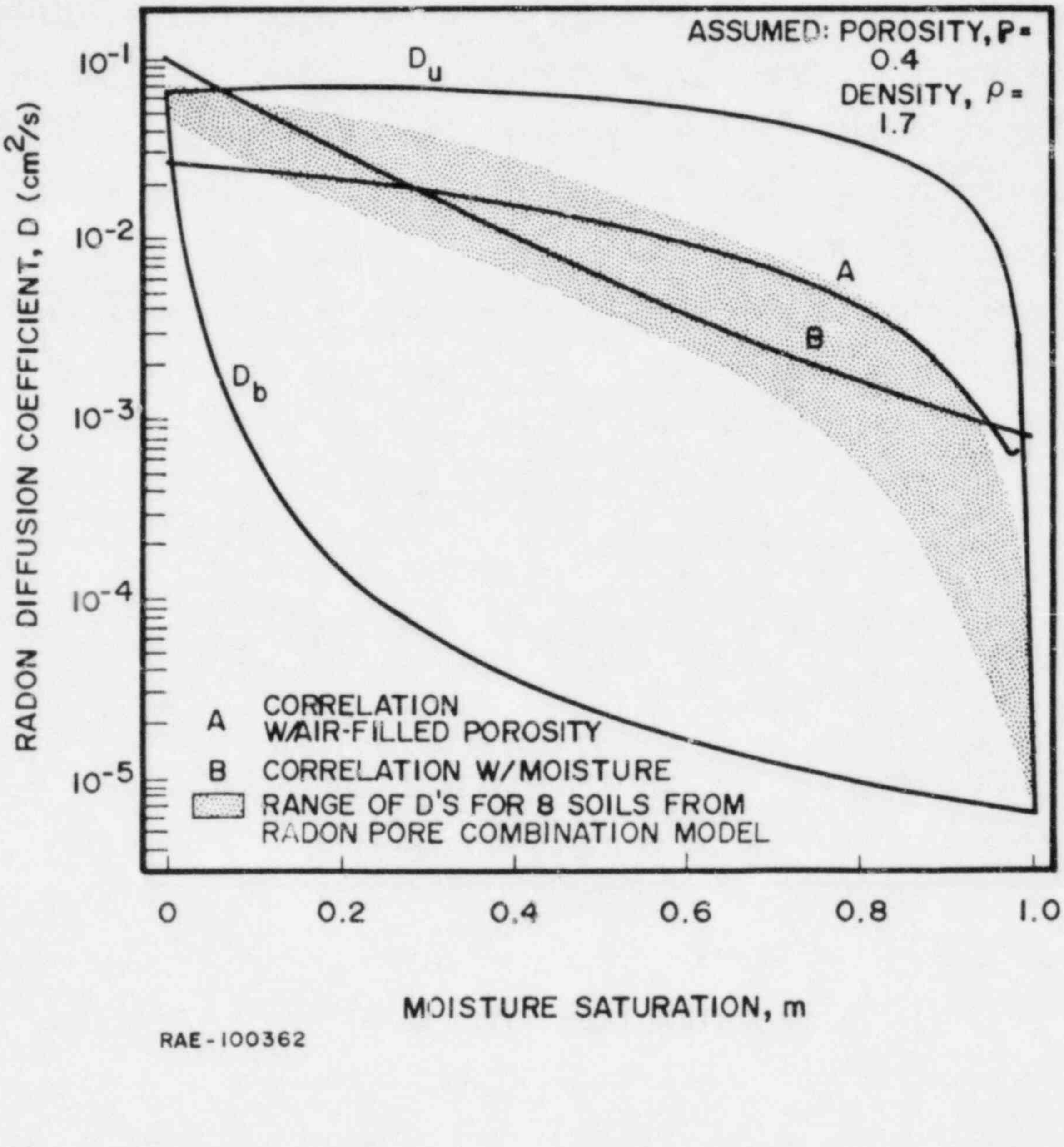

FIGURE 8. COMPARISON OF DIFFUSION COEFFICIENTS FOR WATER-BLOCKED PORES, D_b, AND UNBLOCKED PORES, D_u, WITH EMPIRICAL CORRELATIONS WITH AIR-FILLED POROSITY (A) AND MOISTURE (B), AND WITH THE RANGE OF CALCULATED D'S FOR EIGHT SOILS.

cannot be made with the random pore combination model. However, if the range of coefficients calculated from the random pore combination model illustrated in Figure 7 is assumed to be representative of the soils used in deriving the correlations, agreement would be expected with the correlation curves in Figure 8. As illustrated by the superimposed shaded region, the range of calculated values includes the correlation curves over nearly all of the moisture saturation range. The agreement between the correlations and the model is considered excellent because the correlations are based on measured D's from soils in one study and the model calculations are based on measured pore size distributions from a different set of soils. Since both sets of data in this comparison included a wide variety of sandy, silty, and clay-type soils, it appears likely that the curves illustrated in Figure 8 are sufficiently general to estimate diffusion coefficients for most tailings cover materials.

CONCLUSIONS

The diffusion of radon through porous earthen materials has been characterized by explicit diffusion in both gas and liquid phases. An analytical model based on the moisture content and the pore size distribution allows prediction of the radon diffusion coefficient and its moisture dependence without resorting to radon diffusion measurements. The diffusion coefficients of individual pore segments can be represented by simple functions of their moisture and air contents, which can in turn be used to calculate diffusion coefficients of more complicated pores. The effect of moisture on the radon diffusion coefficients is greater than can be simply explained by porosity considerations, and must rely on the diffusion of radon through short water pathways in some of the interconnecting pores.

The pore size distribution of an earthen material was found to markedly affect both the magnitude and the moisture dependence of its radon diffusion coefficient. A random pore combination model was found to give a suitable representation of the actual soil pore structure for the purpose of predicting radon diffusion coefficients. The pore structure must include all possible combinations of the increments in the pore size distribution, properly weighted, in order to adequately represent the actual pore channels in which radon diffusion occurs.

Radon diffusion coefficients are lower in soils having low median pore diameters than in soils with higher median pore diameters. This dependence on median pore size is greatest at low moisture contents. Soils with extremely uniform pore sizes (geometric standard deviation of pore sizes <2) are predicted to have significantly higher diffusion coefficients than soils with broader pore size distributions.

The random pore combination model was shown to predict radon diffusion coefficients with a relative uncertainty on the order of 50 percent for six soils. Further studies with a greater variety of soils will establish more conclusively the precision and accuracy of this model. The model was also shown to be in excellent agreement with previously reported correlations of diffusion coefficients with moisture and air-filled porosity.

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Radon migration in porous, earthen materials is characterized by diffusion in both the air and water components of the system as well as by the interaction of the radon between the air and water. The size distribution and configuration of the pore spaces and their moisture distributions are key parameters in determining the radon diffusion coefficient for the bulk material. A mathematical model is developed and presented for calculating radon diffusion coefficients soley from the moisture content and pore size distribution of a soil, reducting the need for resorting to radon diffusion measurements. The resulting diffusion coefficients increase with the median pore diameter of the soil and decrease with increasing widths of the pore size distribution. The calculated diffusion coefficients are suitable for use in simple homogeneous-medium diffusion experssions for predicting radon transport and compare well with measured diffusion coefficients and with empirical diffusion coefficient correlations.

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