EFFECT OF TIME DEPENDENT DIFFUSION
ON RADON-222 ATTENUATION EXPERIMENTS

Oktay I. Oztunali, Leslie Skoski
Dames & Moore
20 Haarlem Avenue, White Plains, N.Y. 10603

ABSTRACT

The experimental determination of the attenuation of radon-222 by a given thickness of material or the determination of the diffusion coefficient of the material requires measurements of the steady state radon-222 flux before and after the emplacement of the cover material on top of a known radon-222 source. These measurements are subject to error if a transient rather than a steady state flux is measured. The time required for the transient flux to reach steady state conditions varies depending on the thickness and the diffusion coefficient of the cover material. This paper considers the transient flux through a mathematical solution of the time-dependent diffusion equation. The effect of this time-dependent diffusion on radon-222 attenuation experiments and the necessary formulation for incorporating the time-dependent effects into the experimental results are also presented.

INTRODUCTION

Radon-222 (Rn-222) flux measurements and prediction of the attenuation of the flux by different cover materials has become an important consideration in view of a variety of regulatory positions with regards to the reclamation of uranium mill tailings, mined out phosphate properties, and other areas contaminated with enhanced naturally occurring radionuclides (Ref. 1-3). Accordingly, there has been an increasing number of experiments performed to determine Rn-222 attenuation properties of materials (Refs. 4-6). Previous work performed on Rn-222 behavior in porous media indicates that diffusion theory, with minor modifications, can be applied to the interpretation of experimental results and to the prediction of Rn-222 flux attenuation (Refs. 7-9). A critical component of diffusion theory is the diffusion coefficient, a macroscopic parameter incorporating the effects of several microscopic variables.

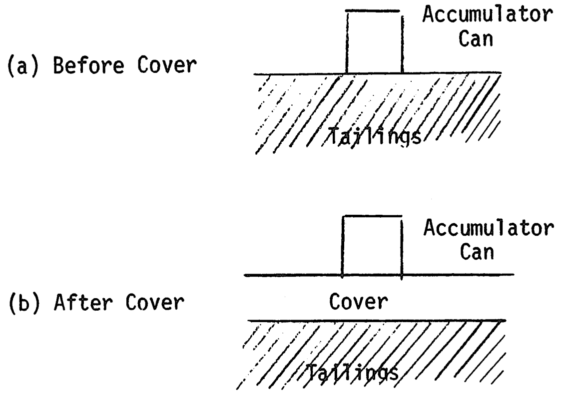

Fig. 1. Experimental Attenuation Geometry

As shown in Fig. 1, a conventional method of determining either attenuation of Rn-222 through a given material or the diffusion coefficient of the material involves measurement of first the Rn-222 flux from a given source (Fig. 1(a)), adding a certain thickness of material, which is assumed to be sourceless, and measuring the Rn-222 flux at a later time (Fig. 1(b)). Frequently, this experiment is repeated for several thicknesses of cover material, and a plot of attenuation vs the thickness generated. This provides a tool for the determination of the diffusion coefficient of the cover material which leads ultimately to the estimation of cover thicknesses for the material.

In the past it has been assumed that the Rn-222 flux measured after the addition of the material on top of the source plane is the steady state flux. This paper quantifies, as a function of the thickness of the material and its diffusion coefficient, the equilibration time or the time required for the flux to approach steady state conditions after the addition of the material. In order to accomplish this, the mathematical solution of the time-dependent diffusion equation is considered.

In sections below, first the theory of the diffusion equation that gives rise to time-dependent solution is considered, and then this solution is examined.

THEORY

The geometry of the Rn-222 diffusion is shown in Fig. 2. At time t=0, a given thickness x_1 of material, which is assumed to be sourceless, with diffusion coefficient D_1 is added on top of the Rn-222 source at x<0 with a steady state flux for t<0 denoted by J_o. The mathematical problem is the solution of the diffusion equation with radioactive decay, i.e.,

$$\left\{ D_1 \frac{\partial^2}{\partial x^2} - \lambda - \frac{\partial}{\partial t} \right\} C(x,t) = 0 \qquad (1)$$

with the conditions

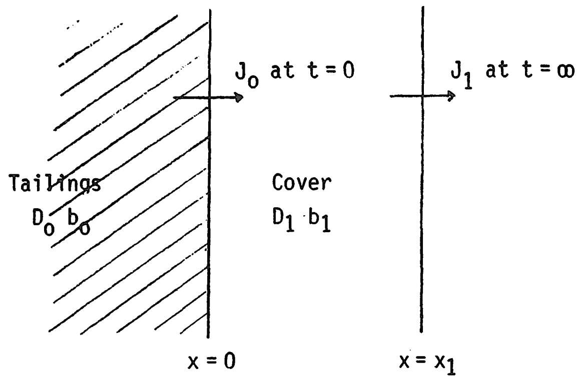

Fig. 2. Radon-222 Diffusion Geometry

$$C(x,t=0) = 0 \tag{2}$$

and

$$-D_1 \frac{\partial}{\partial x} C(x,t) \Big|_{\substack{x=0\\ t=0}} = J_0 \tag{3}$$

and

$$-D_1 \frac{\partial}{\partial x} C(x,t) \Big|_{\substack{x=x_1\\ t\to\infty}} = J_1 \tag{4}$$

with

$$J(x,t) = -D_1 \frac{\partial}{\partial x} C(x,t) \tag{5}$$

where (Ref. 10)

$$J_1 = J_o \exp(-b_1 x_1 h) \tag{6}$$

$$h = 1 - \frac{1}{b_1 x_1} \ln \left\{ 2/ \left(\{ 1 + (\frac{D_o b_o}{D_1 b_1}) \tanh(b_o x_o) \} + \{ 1 - (\frac{D_o b_o}{D_1 b_1}) \tanh(b_o x_o) \} \exp(-2b_1 x_1) \right) \right\} \tag{7}$$

$$b_1 = \sqrt{\lambda / D_1} \tag{8}$$

and where

λ = decay constant of radon-222 (1/sec)

D_1 = Diffusion coefficient of the cover material (cm^2/sec)

$C(x,t)$ = radon-222 concentration (pCi/cm^3)

$J(x,t)$ = radon-222 flux (pCi/cm^2-sec)

x = distance (cm)

t = time (sec)

and where, by convention, the porosity (void fraction) has been incorporated into the diffusion coefficient. This formulation assumes that the existance of a free surface at some positive x would not affect the radon distribution between x=0 and the free surface. This assumption is implicitly made in most of the existing literature; it has not been invalidated by experimental results, and its analogs exist in other disciplines, e.g., treatment of free surfaces via the extrapolation length concept in neutron diffusion theory.

It can be shown that the solution of this set of equations are:

$$C(x,t) = \frac{J_o}{D_1 b_1} \exp(-b_1 x)\ F_-(x,t) + \frac{J_1 e^{b_1 x_1} - J_o}{D_1 b_1} \exp(-b_1 x)\ F_+(x,t) \tag{9}$$

and

$$J(x,t) = J_o \exp(-b_1 x)\ F_+(x,t) + (J_1 e^{b_1 x_1} - J_o) \left\{ e^{-b_1 x}\ F_-(x,t) + \frac{2/b_1}{\sqrt{4\pi D_1 t}} \exp(-\lambda t - \frac{x^2}{4 D_1 t}) \right\} \tag{10}$$

with

$$F_\pm(x,t) = 0.5 \left(\operatorname{erfc} \frac{x - 2D_1 b_1 t}{\sqrt{4 D_1 t}} \pm \exp(2b_1 x) \operatorname{erfc} \frac{x + 2D_1 b_1 t}{\sqrt{4 D_1 t}} \right) \tag{11}$$

and

$$\operatorname{erfc}(x) = 1 - \int_0^x \{ 2/\sqrt{\pi} \} \exp(-x^2) dx \tag{12}$$

It is interesting to note that

$$F_+(0,t) = 1 \tag{13}$$

$$F_-(0,t) = \operatorname{erf}(\sqrt{\lambda t}) \tag{14}$$

and

$$F_\pm(x,0) = 0 \tag{15}$$

$$\lim_{t\to\infty} F_\pm(x,t) = 1 \tag{16}$$

$$\lim_{x\to\infty} F_\pm(x,t) = 0 \tag{17}$$

These conditions verify that the above initial and boundary conditions are satisfied. It is also interesting to note that the second term of the flux $J(x,t)$ drops out in the extremely thick tailings case (i.e., tanh $D_o b_o$ = 1, with $D_o b_o = D_1 b_1$).

RESULTS AND DISCUSSION

Equation (10) in this extreme case can be restated as follows:

$$\frac{J(x,t)}{J_o \exp(-b_1 x_1)} = F_+(x_1,t) \tag{18}$$

This equation was utilized to determine the values of time for which $F_+(x,t)$ is 0.5 for several selected values of x_1 and D_1. The hours required to achieve the mid-point equilibrium are given below.

Hours to Mid-point Equilibrium

Diffusion Coefficient	Thickness x_1		
D_1 (cm^2/sec)	1 ft	2 ft	3 ft
0.01	14.13	37.83	64.14
0.003	33.44	81.89	132.89
0.001	68.53	156.92	247.47
0.0003	141.31	306.94	474.17
0.0001	262.25	551.94	842.50

Clearly, a correction factor must be incorporated into the Rn-222 attenuation experiment results depending on the waiting time utilized after the emplacement of the cover material in order to account for the non-steady state conditions of the flux.

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