

TABLE 1. Parameters of Radon Diffusion in Low-Density Polyethylene

Method	D, cm <sup>2</sup> /sec	P, cm <sup>2</sup> /sec·Pa	K <sub>p</sub> , cm <sup>3</sup> /Pa
Integral	(5.2±0.4)·10 <sup>-8</sup>	(4.4±0.4)·10 <sup>-13</sup>	(0.85±0.02)·10 <sup>-5</sup>
Differential	(3.2±0.1)·10 <sup>-8</sup>	(2.6±0.5)·10 <sup>-13</sup>	(0.82±0.05)·10 <sup>-5</sup>

The plot by Eq. (5) for long durations attains the constant quantity  $I_{\infty} = (DSC_0)/H$ , from which the penetrability constant can be found as

$$P = \frac{Q_{\infty} H}{S p} \quad (6)$$

The diffusion constant can be determined from the time for attaining a flow rate half that in the stationary state (i.e.,  $\tau_{1/2}$  is a time at which  $J_1/J_0 = 0.5$ ):

$$D = \frac{H^2}{7.2 \tau_{1/2}^2} \quad (7)$$

Study of Radon Diffusion in Polyethylene. The methods proposed were used to study the diffusion of the radioactive inert gas radon (<sup>222</sup>Rn) in low-density (57% crystallinity) polyethylene films. The typical experimental plots are given in Fig. 3. The evaluations of the parameters of radon diffusion in polyethylene at the 90% significance level (temperature 18°C), made in the integral (three specimens, seven measurements) and differential (four specimens, seven measurements) methods, are given in Table 1.

The referred values agree well with the theoretical ones based on physicochemical properties of radon and with the diffusion parameters of other gases in low-density polyethylene [3].

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#### EXPERIMENTAL STUDIES OF RADIOACTIVE GAS DIFFUSION IN SOLIDS.

#### II. EVALUATION OF RADIOACTIVE DECAY AND GROWTH IN THE PENETRANCE METHOD

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The phenomenological theory of the radiochemical penetrance method is discussed with due regard for radioactive decay and growth. Expressions are given for time dependence of flow, amount of substance diffused, and concentration distribution across the membrane thickness under conditions of gradual radiochemical conversions. It is shown that the time lag may diminish (on radioactive decay) or increase (due to active deposition). Equations are proposed for calculating diffusion constants of gases labelled with short-lived isotopes having complex decay characteristics (e.g., of radon) from the experimental data obtained by the penetrance method.

Processing of experimental data obtained in the radiochemical version of the penetrance method often makes it necessary to take account of complex processes of radioactive decay and

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growth accompanying diffusion of the test isotope. For instance, when radon is used as a penetrant, its radioactive decay characteristics (e.g., various types of radiations, complex decay and growth processes, etc.) renders interpretation of kinetic curves quite difficult. In fact, in the integral version of the penetrance method the activity in the closed receiver will increase not only due to increased concentration of the gas diffused through the membrane, but also due to a multistage process of active deposit formation. In the differential version flow of radioactive filial atoms may also occur in addition to the flow of radon atoms. The fall in radon concentration due to radioactive decay should be given due consideration when the diffusion constants are low or the membrane is very thick.

This work is undertaken to analyze the solutions of diffusion equations in the penetrance method involving radioactive decay and growth processes. Attention is focused primarily on the deduction of mathematical equations necessary for processing experimental data from the diffusion study of gases labelled with short-lived isotopes or with isotopes having complex decay characteristics. The problems of concurrent use of emanation and penetrance methods are discussed.

Note that, in addition to their direct use for radiochemical purposes, the expressions derived can also be used for processing data on diffusion accompanied by first-order chemical reactions and consecutive reactions, as also for the study of diffusion in defective media (diffusion with constant or temporal retention).

Solution of Diffusion Equations Involving Radioactive Decay. Gas transport through thin membranes with the occurrence of first-order chemical reactions or radioactive decay can be described by solving the equations for the first and second laws of Fick as follows:

$$Q = -DS \left( \frac{\partial C^*}{\partial x} \right)_{x=0}, \quad (1)$$

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

under the boundary conditions

$$C^*(0, t) = 0; \quad C^*(H, t) = C_0^*; \quad C^*(x, 0) = 0, \quad (3)$$

where  $C^*(x, t)$  is the concentration distribution of the diffusing radioactive gas across the membrane thickness;  $C_0^*$  is the equilibrium gas solubility in the surface layer of the membrane;  $D$  is the diffusion constant;  $S$  is the membrane area;  $H$  is the membrane thickness;  $x$  is the coordinate;  $t$  is the time;  $Q$  is the flow on the outer surface of the membrane; and  $\lambda$  is the radioactive decay constant (or the rate constant of the first-order chemical reaction).

Putting  $C^* = C \cdot \exp\{-\lambda t\}$  [1] in Eq. (2) we obtain the latter in the usual form:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2}. \quad (4)$$

Let decay occur only in the membrane. Such a situation can be attained experimentally, for instance, by sustaining a constant gas concentration in the reservoir with the help of radium-radon equilibrium. Then  $C_0^* = C_0$  and the general solution can be put in the form [2]

$$C^* = \lambda \int_0^t e^{-\lambda t} \cdot C \cdot dt + C e^{-\lambda t}, \quad (5)$$

where  $C$  is the solution for the respective equations in the absence of radioactive decay:

$$C(x, t) = \frac{C_0 x}{H} + \frac{2C_0}{\pi} \sum_{n=1}^{\infty} \frac{(-1)^n}{n} \sin \frac{n\pi x}{H} \exp\{-n^2 B t\}, \quad (6)$$

where  $B = \pi^2 D / H^2$ .

The change of gas concentration distribution across the membrane thickness with time is

$$C^*(t, x) = \frac{C_0 x}{H} + \frac{2C_0 \lambda}{\pi} \sum_{n=1}^{\infty} \frac{(-1)^n \sin \frac{n\pi x}{H}}{n(n^2 B + \lambda_0)} [1 - \exp\{-(n^2 B + \lambda) t\}] + 2 \frac{C_0}{\pi} \sum_{n=1}^{\infty} \frac{(-1)^n}{n} \sin \frac{n\pi x}{H} \exp[-(n^2 B + \lambda) t]. \quad (7)$$

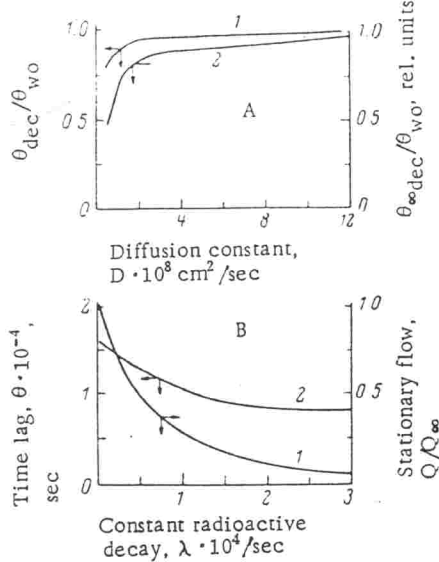


Fig. 1. Effect of radioactive decay on diffusion time lag and stationary penetrating flow ( $H = 0.1$  cm). A) Dependence of  $\theta_{\text{dec}}/\theta_{\text{wo}}$  and  $Q_{\text{dec}}/Q_{\text{wo}}$  on diffusion constant at a fixed level of constant decay ( $\lambda = 2.1 \cdot 10^{-6}/\text{sec}$ ): 1) time lag during decay and 2) stationary penetrating flow during decay. B) Dependence of  $\theta_{\text{dec}}/\theta_{\text{wo}}$  and  $Q_{\text{dec}}/Q_{\text{wo}}$  on constant radioactive decay at a fixed diffusion constant ( $D = 10^{-7}$   $\text{cm}^2/\text{sec}$  and  $H = 0.1$  cm): 1) stationary flow and 2) time lag.

The flow, i.e., the amount of the substance passing at a point  $x = 0$  in unit time through the membrane of area  $S$ , is

$$Q(t) = \frac{DSC_0}{H} \left[ 1 + 2 \sum_{n=1}^{\infty} (-1)^n \exp\{-(n^2B + \lambda)t\} + 2\lambda \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2B + \lambda} [1 - \exp\{-(n^2B + \lambda)t\}] \right]. \quad (8)$$

The volume of the gas passing through the membrane in time  $t$  is

$$q(t) = \frac{DSC_0}{H} \left( t + 2 \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2B + \lambda} [1 - \exp\{-(n^2B + \lambda)t\}] + 2\lambda t \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2B + \lambda} - 2\lambda \sum_{n=1}^{\infty} \frac{(-1)^n}{(n^2B + \lambda)^2} [1 - \exp\{-(n^2B + \lambda)t\}] \right). \quad (9)$$

For long durations the plot by Eq. (9) is a straight line describing a stationary state and the segment intercepted by the extension of the rectilinear segment on the time axis (the so-called "time lag") is

$$\theta = \frac{2 \sum_{n=1}^{\infty} (-1)^n \frac{n^2B}{(n^2B + \lambda)^2}}{1 + 2\lambda \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2B + \lambda}} = \frac{2}{H} \sqrt{\frac{D}{\lambda}} \text{Sh} \sqrt{\frac{\lambda}{D}} H \sum_{n=1}^{\infty} \frac{(-1)^n n^2B}{(n^2B + \lambda)^2}. \quad (10)$$

The stationary flow with  $t \rightarrow \infty$  is described by the expression

$$Q_{\infty} = \frac{DSC_0}{H} \left[ 1 + 2\lambda \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2B + \lambda} \right]. \quad (11)$$

It can be proved readily that with  $\lambda \rightarrow 0$  expressions (7)-(11) become the usual expressions for diffusion of stable isotopes.

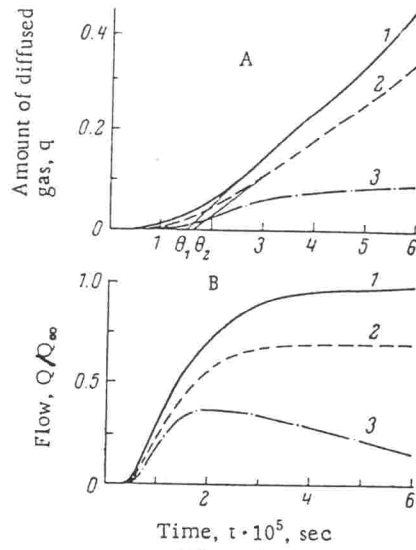


Fig. 2

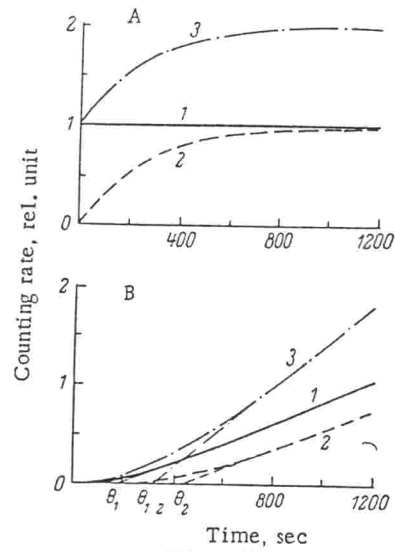


Fig. 3

Fig. 2. Effect of radioactive decay on kinetic curves in integral and differential versions of penetrance method ( $D = 10^{-8}$   $\text{cm}^2/\text{sec}$ ,  $\lambda = 2.1 \cdot 10^{-6}/\text{sec}$ , and  $H = 0.1$  cm). A) Amount of substance diffused through the membrane as a function of time. B) Material flow through the membrane with time. 1) Without decay ( $\theta = 1.667 \cdot 10^5$  sec); 2) decay in the membrane ( $\theta = 1.471 \cdot 10^5$  sec); and 3) decay in the reservoir and membrane.

Fig. 3. Effect of radioactive decay products of a diffusing isotope on kinetic curves in the integral version of the penetrance method.  $D = 10^{-7}$   $\text{cm}^2/\text{sec}$ ;  $H = 0.01$  cm;  $\theta = 167$  sec;  $\lambda_1 = 2.08 \cdot 10^{-6}/\text{sec}$ ; and  $\lambda_2 = 3.79 \cdot 10^{-3}/\text{sec}$ . A) Activity change in the reservoir and B) activity change in the receiver; 1) activity of the parent isotope  $A_1$  (radon); 2) activity of the filial isotope  $A_2$  (RaA); and 3) total activity.

Figure 1 demonstrates the effect of radioactive decay on the time lag and stationary flow. Both stationary flow and time lag diminish when radioactive decay occurs in the membrane. The distortion of kinetic curves in the integral and differential versions of the penetrance method is shown in Fig. 2. It is evident that decay may cause significant distortion of the shape of the experimental plot.

If the gas in the reservoir also decays,  $C_0^\circ = C_0 \exp \{-\lambda t\}$ , where  $C_0^\circ$  is the gas concentration in the surface layer of the membrane at  $t = 0$ . Decay in the reservoir does not alter the time lag.

Penetrability with the Radioactive Gas Sources Present in the Membrane. Let us now refer to a more general case, viz., penetrability when the sources of the radioactive gas occur in the membrane itself. A similar situation arises, for instance, when emanation and penetrance methods are used simultaneously.

Let the radioactive gas with a constant decay  $\lambda_2$  be distributed initially across the membrane thickness obeying a certain function  $C(x, 0) = f(x)$ . The source of the radioactive gas with a power  $F$  is active in the membrane from the instant  $t = 0$ . Since our interests rest on the emanation method, we shall limit ourselves to a discussion of the case  $F = \lambda_1 C_1$ , where  $\lambda_1$  is the constant decay and  $C_1$  is the concentration of the parent isotope (e.g.,  $^{226}\text{Ra}$ ). Let at a later point of time a radioactive gas concentration  $C_2^H$  be created on one membrane surface and a concentration  $C_2^0$  on the other surface. Then our task is reduced to the solution of the diffusion equation taking account of the source and the flow as below:

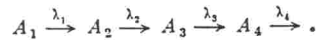
$$\frac{\partial C_2}{\partial t} = D \frac{\partial^2 C_2}{\partial x^2} + \lambda_1 C_1 - \lambda_2 C_2. \quad (12)$$

Solving Eq. (12) by standard methods of mathematical physics [3] we obtain the emanation concentration distribution across the membrane thickness:

$$\begin{aligned}
 C(x, t) = & \frac{\lambda_1 C_1}{\lambda_2} \left[ 1 - \frac{\operatorname{ch} \sqrt{\frac{\lambda_2}{D}} \left( x - \frac{H}{2} \right)}{\operatorname{ch} \frac{H}{2} \sqrt{\frac{\lambda_2}{D}}} \right] - \frac{4\lambda_1 C_1}{\pi} \sum_{n=0}^{\infty} \frac{\sin \frac{(2n+1)\pi x}{H} \exp \{-[(2n+1)^2 B + \lambda_2] t\}}{(2n+1) [(2n+1)^2 B + \lambda_2]} + \\
 & + \frac{C_2^0 \operatorname{Sh} \sqrt{\frac{\lambda_2}{D}} (H-x) + C_2^H \operatorname{Sh} \sqrt{\frac{\lambda_2}{D}} x}{\operatorname{Sh} \sqrt{\frac{\lambda_2}{D}} H} + \frac{2D\pi}{H^2} \sum_{n=1}^{\infty} n \frac{(-1)^n C_2^H - C_2^0}{n^2 B + \lambda_2} \sin \frac{n\pi x}{H} \exp \{-[(2n+1)^2 B + \lambda_2] t\} + \\
 & + \frac{2}{H} \sum_{n=1}^{\infty} \sin \frac{n\pi x}{H} \exp \{-[n^2 B + \lambda_2] t\} \int_0^H f(x) \sin \frac{n\pi x}{H} dx. \quad (13)
 \end{aligned}$$

Equation (13) is a fairly general one permitting one to arrive at equations used both in the emanation method and in the penetrance method with due regard for radioactive decay.

Solution of Diffusion Equations for Gradual Radiochemical Conversions. Let a radioactive gas  $A_1$  decay consecutively to a series of radioisotopes following the scheme



Each radioisotope is characterized by a proper value of constant decay and diffusion constant ( $\lambda_i, D_i$ ).

Integral Version of Penetrance Method. For simplicity let us assume that the diffusion constant of the decay products is much lower than the diffusion constant of the gas. Then the activity in the closed chamber of the receiver will increase due to gas diffusion and to accumulation of products of gas decay in the receiver. Further, suppose the decrease in the number of gas atoms in the receiver due to gas decay is negligible and there is no activity in the receiver at the starting point of time. Then the change in the number of atoms of the  $i$ -th isotope of the series ( $i > 1$ ) is described by the differential equation

$$\frac{\partial q_i}{\partial t} = \lambda_{i-1} q_{i-1} - \lambda_i q_i, \quad (14)$$

the general solution of which is

$$q_i = e^{-\lambda_i t} \left[ q_0 + \int_0^t \lambda_{i-1} q_{i-1}(t) e^{-\lambda_i t} dt \right]. \quad (15)$$

In our case  $q_0$  and  $i = 1$ , and  $q_i$  is given by the equation [2]

$$q_i(t) = C_0 D S \left[ \frac{t}{H} - \frac{H}{6D} \sum_{n=1}^{\infty} \left( 1 + \frac{12}{\pi^2} \cdot \frac{(-1)^n}{n^2} \exp \{-n^2 B t\} \right) \right]. \quad (16)$$

Putting (16) in (15) and integrating, we obtain (taking account of Henry's law) the following for the rates of counting of each isotope in the receiver:

$$I_1 = a_1 k_1 P \frac{S}{H} \frac{T}{T_0} p_0 \left[ t - \frac{H^2}{6D} - \frac{2H^2}{\pi^2 D} \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} \exp \{-n^2 B t\} \right], \quad (17)$$

$$I_2 = a_1 k_2 P \frac{S}{H} \frac{T}{T_0} p_0 \left[ t - \left( \frac{1}{\lambda_2} + \frac{H^2}{6D} \right) (1 - \exp \{-\lambda_2 t\}) \right] - \frac{2H^2 \lambda_2}{\pi^2 D} \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} \cdot \frac{\exp \{-n^2 B t\} - \exp \{-\lambda_2 t\}}{\lambda_2 - n^2 B} \quad (18)$$

$$\begin{aligned}
 I_3 = & a_1 k_3 P \frac{S}{H} \frac{T}{T_0} p_0 \left\{ t - \left( \frac{1}{\lambda_2} + \frac{1}{\lambda_3} + \frac{H^2}{6D} \right) [1 - \exp \{-\lambda_3 t\}] + \frac{\lambda_3}{\lambda_3 - \lambda_2} \left( \frac{1}{\lambda_2} + \frac{H^2}{6D} \right) [\exp \{-\lambda_2 t\} - \exp \{-\lambda_3 t\}] + \right. \\
 & \left. + \frac{12H^2}{\pi^2 D} \lambda_2 \lambda_3 \sum_{n=1}^{\infty} \frac{(-1)^n}{n} \left[ \frac{\exp \{-\lambda_2 t\} - \exp \{-\lambda_3 t\}}{(\lambda_2 - n^2 B)(\lambda_3 - \lambda_2)} - \frac{\exp \{-n^2 B t\} - \exp \{-\lambda_3 t\}}{(\lambda_2 - n^2 B)(\lambda_3 - n^2 B)} \right] \right\} \quad (19)
 \end{aligned}$$

$$\begin{aligned}
I_4 = & a_1 k_4 P \frac{S}{H} \cdot \frac{T}{T_0} p_0 \left\{ t - \left( \frac{1}{\lambda_2} + \frac{1}{\lambda_3} + \frac{1}{\lambda_4} + \frac{H^2}{6D} \right) [1 - \exp(-\lambda_4 t)] \right. \\
& + \frac{\lambda_4}{\lambda_4 + \lambda_3} \left[ \frac{1}{\lambda_2} + \frac{1}{\lambda_3} + \frac{\lambda_3}{\lambda_2(\lambda_2 - \lambda_3)} + \frac{H^2}{6D} + \frac{H^2 \lambda_3}{6D(\lambda_2 - \lambda_3)} \right] [\exp(-\lambda_3 t) - \exp(-\lambda_4 t)] \\
& - \frac{\lambda_3 \lambda_4}{(\lambda_3 - \lambda_2)(\lambda_4 - \lambda_2)} \left[ \frac{1}{\lambda_2} + \frac{H^2}{6D} \right] [\exp(-\lambda_4 t) - \exp(-\lambda_2 t)] - \frac{2H^2}{6D} \frac{\lambda_2 \lambda_3 \lambda_4}{(\lambda_3 - \lambda_2)(\lambda_4 - \lambda_2)} \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} \cdot \frac{\exp(-\lambda_4 t) - \exp(-\lambda_2 t)}{n^2 B - \lambda_2} \\
& - \frac{2H^2}{\pi^2 D} \frac{\lambda_2 \lambda_3 \lambda_4}{\lambda_4 - \lambda_3} \left[ \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} \cdot \frac{1}{(\lambda_2 - n^2 B)(\lambda_3 - \lambda_2)} \right. \\
& \left. - \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} \cdot \frac{1}{(\lambda_2 - n^2 B)(\lambda_3 - n^2 B)} \right] [\exp(-\lambda_3 t) - \exp(-\lambda_4 t)] \\
& \left. - \frac{2H^2}{\pi^2 D} \lambda_2 \lambda_3 \lambda_4 \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} \cdot \frac{\exp(-n^2 B t) - \exp(-\lambda_4 t)}{(\lambda_2 - n^2 B)(\lambda_3 - n^2 B)(\lambda_4 - n^2 B)} \right\}. \quad (20)
\end{aligned}$$

where P is the penetrability constant;  $\alpha_1$  is the specific gas activity in the reservoir; T is the temperature ( $^{\circ}$ K);  $T_0$  and  $p_0$  are the normal temperature and pressure, respectively; and  $k_1$  and  $k_2$  are the counting coefficients for each isotope.

For calculating radioactive gas decay in the reservoir the right parts of Eqs. (17)-(20) should be multiplied by  $\exp\{-\lambda_1 t\}$ .

The counting rate-time plot for each individual member of the series for long durations becomes straight and the slope of the individual straight lines for each member of the series is the same (provided the counting coefficients are the same). The time lags for each individual isotope are respectively

$$\theta_1 = \frac{H^2}{6D}, \quad (21)$$

$$\theta_2 = \frac{H^2}{6D} + \frac{1}{\lambda_2}, \quad (22)$$

$$\theta_3 = \frac{H^2}{6D} + \frac{1}{\lambda_2} + \frac{1}{\lambda_3}, \quad (23)$$

$$\theta_4 = \frac{H^2}{6D} + \frac{1}{\lambda_2} + \frac{1}{\lambda_3} + \frac{1}{\lambda_4} \quad (24)$$

If the activities of the isotopes  $A_1$  and  $A_2$  are recorded simultaneously, then

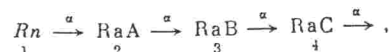
$$\theta_{1+2} = \frac{H^2}{6D} + \frac{1}{2\lambda_2}. \quad (25)$$

If the activities of the isotopes  $A_1$ ,  $A_2$ , and  $A_3$  are recorded, then

$$\theta_{1+2+3} = \frac{H^2}{6D} + \frac{2}{3} \cdot \frac{1}{\lambda_2} + \frac{1}{3\lambda_3} \quad (26)$$

and so on. It is evident from the referred equations that the time lag increases due to the effect of decay products.

Let us now consider radon diffusion. It is well known that radon decays forming a series of decay products (active deposition). The decay products are  $\alpha$ -,  $\beta$ -, and  $\gamma$ -particle radiators. Since  $\alpha$ -particle radiation detectors are generally used for radon recording, the basis scheme of radon decay (ignoring chain branching and long-lived members of the series) can be depicted as



where  $\lambda_1 = 2.08 \cdot 10^{-6}$ /sec;  $\lambda_2 = 3.79 \cdot 10^{-3}$ /sec;  $\lambda_3 = 4.31 \cdot 10^{-4}$ /sec; and  $\lambda_4 = 5.86 \cdot 10^{-4}$  sec.

The total activity of the substances in the receiver will comprise the  $\alpha$ -activity of the three isotopes  $A_1$ ,  $A_2$ , and  $A_3$  and therefore the diffusion constant of radon should be calculated by the equation

$$D = \frac{H^2}{6\theta_{1+2+4} - \frac{2}{3\lambda_2} - \frac{1}{3\lambda_3} - \frac{1}{3\lambda_4}} \quad (27)$$

As an example we calculated the curves for activity change in the reservoir (Fig. 3a) and receiver (Fig. 3b) for a case of radon diffusion at the time lag  $\theta = 167$  sec. A radioactive equilibrium is established between the isotopes  $A_1$  and  $A_2$  within the time span of the experiment on gas penetrance in the reservoir. It is evident from Fig. 3 that the formation of the isotope  $A_2$  exerts much influence on the time-bound change of the activity in the receiver (the penetrability constant rose twofold and the time lag by 70%). The activity of the isotope  $A_4$  is negligible for the referred time lags.

Let us now consider the relative share of the active deposition in the time lags in the case of radon diffusion in membranes with different diffusion constants ( $H = 0.1$  cm). Then for  $D = 10^{-7}$  cm<sup>2</sup>/sec,  $\theta_1 = 167$  sec,  $\theta_2 = 430$  sec,  $\theta_{1+2} = 298$  sec, and  $\theta_{1+2+3+4} = 1688$ , i.e., the time lag rose by one order. For  $D = 10^{-8}$  cm<sup>2</sup>/sec,  $\theta_1 = 1667$  sec,  $\theta_{1+2} = 1798$  sec, and  $\theta_{1+2+4} = 3188$ , i.e.,  $\theta$  rose almost twofold. For  $D = 10^{-9}$  cm<sup>2</sup>/sec,  $\theta_1 = 16,667$  sec,  $\theta_{1+2} = 16,798$  sec, and  $\theta_{1+2+4} = 18,188$  sec, i.e.,  $\theta$  rose approximately by 9%.

The example shows that active deposition exerts much influence on time lag. However, the higher the diffusional time lag, the weaker this influence. Therefore, the membrane thickness should be so chosen in the experiments that this influence could be ignored.

Differential Version. If the decay products of the parent isotope are capable of diffusing with the same diffusion constant as that of the parent isotope, the activity recorded by the differential detector will be the sum of the flow of the parent isotope decayed during its passage through the membrane and the flow of the filial isotope formed in the membrane.

The flow of the parent isotope through the membrane in combination with radioactive decay can be determined by Eq. (8). The flow of the daughter isotope is

$$Q_2 = DSC_0\lambda_1 \left\{ \frac{1}{\lambda_2 H} [1 - \exp(-\lambda_2 t)] + \frac{2}{H} \frac{\lambda_1}{\lambda_2} \sum_{n=1}^{\infty} \frac{(-1)^n [1 - \exp(-\lambda_2 t)]}{n^2 B + \lambda_1} + \frac{2\lambda_1}{H} \sum_{n=1}^{\infty} \frac{(-1)^n \exp[-(n^2 B + \lambda_1)t] - \exp(-\lambda_2 t)}{(n^2 B + \lambda_1)(n^2 B + \lambda_1 - \lambda_2)} - \frac{2}{H} \sum_{n=1}^{\infty} \frac{(-1)^n [\exp(-n^2 B + \lambda_1)t] - \exp(-\lambda_2 t)}{n^2 B + \lambda_1 - \lambda_2} \right\} \quad (28)$$

In the stationary regime

$$Q_2^{(\infty)} = \frac{DSC_0}{H} \cdot \frac{\lambda_1}{\lambda_2} \left[ 1 + 2\lambda_1 \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2 B + \lambda_1} \right] \quad (29)$$

The total flow in the stationary state is

$$Q_{1+2}^{(\infty)} = \frac{DSC_0}{H} \cdot \frac{\lambda_1 + \lambda_2}{\lambda_2} \left[ 1 + 2\lambda_1 \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2 B + \lambda_1} \right] \quad (30)$$

i.e., the stationary flow increases.

Thus, we have shown that the time lag may decrease (on radioactive decay of radon) or increase (due to active deposition). Decay and growth distort the shape of the experimental plots and this may cause errors in the calculation of diffusion constants. The referred factors should be taken into account in diffusion study employing the penetrance method.

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