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RADIATION HAZARD RESULTING FROM TRITIUM DIFFUSION IN GLOVE BOX OPERATIONS

by

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ABSTRACT

Tritium exposure to personnel handling tritium compounds in glove boxes can be significantly reduced by the wearing of surgeon's gloves while working in the dry-box gloves. To investigate this reduction quantitatively and to determine criteria for the frequency of glove change, mathematical analyses of the diffusion of HTO through dry-box rubber gloves with and without surgeon's gloves are made. The relative amounts of HTO absorbed by the worker as a function of time in the two cases are then compared. For tritium concentrations of one curie per cubic meter in the glove-box atmosphere, a change of surgeon's gloves every 15 to 20 minutes is recommended as a safe working procedure.

ACKNOWLEDGEMENT

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It is generally recognized that the primary radiation hazard to personnel handling tritium compounds in glove boxes is from the tritium (generally in the form of HTO) which diffuses through the rubber gloves of the glove box. Since this tritium is readily absorbed by the skin, the worker's arms are generally protected by heavy sleeves, and the hands, which are in more intimate contact, are best protected by surgeon's gloves.

To obtain quantitative information of both the value of the surgeon's gloves and the effectiveness of frequently changing such gloves, the rate of diffusion of HTO through both sets of gloves as a function of time was calculated. In solving this problem the following assumptions were made:

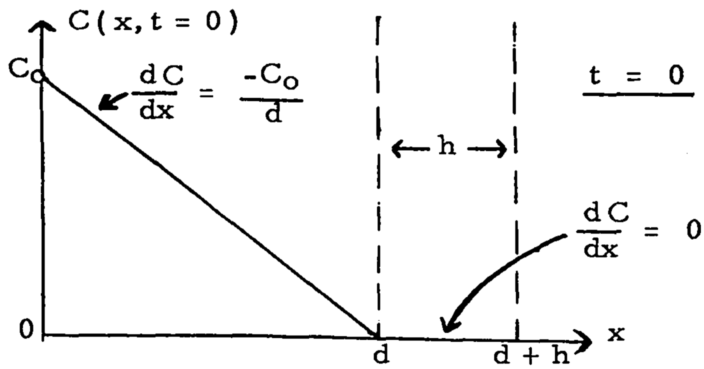
1. Both the permanent and the surgeon's gloves have the same diffusion constant D.

2. This diffusion constant D is, to a first approximation, independent of both the concentration of HTO in the rubber and the temperature of the gloves.

3. The glove material is considered to be planar.

4. The dry-box is an infinite source of HTO at a fixed concentration, and the worker is an infinite sink for HTO.

Initially the permanent gloves are assumed to be in a steady-state condition with a constant concentration gradient throughout; the surgeon's gloves are of course initially at zero HTO concentration throughout. At $t = 0$, therefore, the concentration as a function of distance is as follows (Fig. 1):



d = thickness of the permanent glove
h = thickness of the surgeon's glove

Fig. 1 Initial Tritium Distribution

At $t = \infty$ we again obtain a steady-state situation (Fig. 2), but

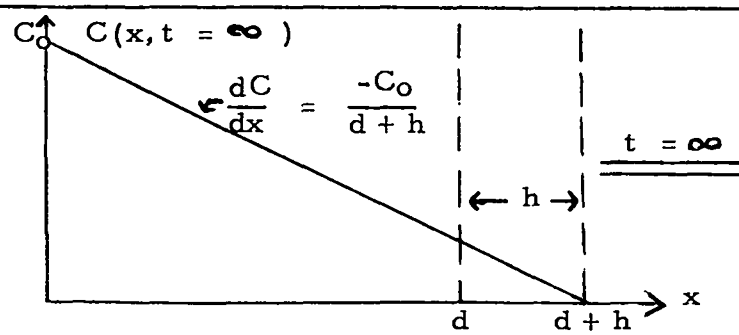


Fig. 2 Steady-State Tritium Distribution

with the concentration gradient in the interval 0 to d reduced somewhat due to the increased effective thickness of the rubber. We always assume the boundary conditions, $C(0,t) = C_o$ and $C(d+h, t) = 0$.

With the addition of tritium-free surgeon's gloves at t = 0, ours becomes a non-steady-state diffusion problem. Accordingly we solve Fick's Law, which for constant D is

$$\frac{\partial C(x,t)}{\partial t} = D \frac{\partial^2 C(x,t)}{\partial x^2},$$

subject to the boundary conditions

$$C(x,0) = \left\{ \begin{array}{ll} (C_o - \frac{C_o x}{d}) & \text{for } 0 \leq x \leq d \\ 0 & \text{for } d \leq x \leq d+h \end{array} \right\} \equiv f_o(x)$$

$$C(d+h, t) = 0; \qquad C(0,t) = C_o.$$

We solve the problem by a method analogous to that found in Carslaw and Jaeger[1] by letting

$$C(x,t) = C_1(x) + C_2(x,t),$$

where C_1 and C_2 satisfy

$$\left\{ \begin{array}{l} \frac{d^2 C_1}{dx^2} = 0 \quad (0 \leq x \leq d+h) \quad \text{(steady-state case)} \\ C_1(0) = C_o; \qquad C_1(d+h) = 0 \quad \text{for all } t \end{array} \right.$$

$$\left\{ \begin{array}{llll} \frac{\partial C_2(x,t)}{\partial t} & = & D\frac{\partial^2 C_2(x,t)}{\partial x^2} & (1) \\ C_2(0,t) & = & C_2(d+h,t) = 0 & (2) \\ C_2(x,0) & = & f_o(x) - C_1(x). & (3) \end{array} \right.$$

The latter equation assures that $C(x,0) = C_1(x) + C_2(x,0) = f_o(x)$.

The solution for $C_1(x)$ is readily obtained by double integration giving

$$C_1(x) = C_o\,(1 - x/d+h),$$

which is our steady-state solution at $t = \infty$.

To solve for $C_2(x,t)$ we let $C_2(x,t) = X(x)T(t)$. Dividing equation (1) by $X(x)T(t)$ we get

$$\frac{1}{D}\,\frac{1}{T(t)}\,\frac{\partial T(t)}{\partial t} = \frac{1}{X(x)}\,\frac{\partial^2 X(x)}{\partial x^2} = -\alpha^2,$$

where α^2 is a constant since one side of the equation is a function of x only and the other of t only. The solutions are

$$X(x) = \sin \alpha x$$
$$T(t) = e^{-\alpha^2 Dt}\quad.$$

Any sum of solutions is also a solution ; and by the boundary conditions on $C_2(x,t)$, equation (2), we have that $\alpha_n = \frac{n\pi}{d+h}$, $n = 1, 2, 3, \ldots$ for the permissible values of α. Thus

$$C_2(x,t) = X(x)T(t) = \sum_{n=1}^{\infty} b_n \sin \alpha_n x e^{-\alpha_n^2 Dt}$$
$$= \sum_{n=1}^{\infty} b_n \sin\left(\frac{n\pi x}{d+h}\right) e^{\frac{-n^2\pi^2 Dt}{(d+h)^2}}\quad.$$

But $C_2(x,o) = \left[f_o(x) - C_1(x)\right] = \sum_{1}^{\infty} b_n \sin\left(\frac{n\pi x}{d+h}\right)$;

or, using the orthogonality of sine functions over a periodic interval,

$$b_n = \frac{2}{d+h}\int_0^{d+h}\left[f_o(x') - C_1(x')\right]\sin\left(\frac{n\pi x'}{d+h}\right)\,dx'\ .$$

Therefore, our general solution for $C(x,t)$ is

$$C(x,t) = C_o\left(1-\frac{x}{d+h}\right) + \sum_{n=1}^{\infty}\left\{\frac{2}{d+h}\int_0^{d+h}\left[f_o(x') - C_1(x')\right]\sin\frac{n\pi x'}{d+h}dx'\right\} \bullet$$

$$\left\{\sin\frac{n\pi x}{d+h}\, e^{\frac{-n^2\pi^2 Dt}{(d+h)^2}}\right\};$$

or after explicit evaluation of the integrals to find the b_n,

$$C(x,t) =$$

$$C_o\left(1-\frac{x}{d+h}\right) - \frac{2C_o(d+h)}{\pi^2 d}\sum_1^{\infty}\frac{1}{n^2}\sin\left(\frac{n\pi d}{d+h}\right)\sin\left(\frac{n\pi x}{d+h}\right)\left(e^{\frac{-Dn^2\pi^2 t}{(d+h)^2}}\right) .$$

Knowing $C(x,t)$ we now find the total amount $Q(t)$ of HTO which has diffused through the gloves by integrating the instantaneous expression

$$\frac{dQ(t)}{dt} = -D\frac{\partial C(x,t)}{\partial x} , \quad \text{or}$$

$$Q(t) = -D\int_0^t \frac{\partial C(x,t')}{\partial x}\bigg|_{x=d+h} dt' ,$$

where we evaluate $\partial C/\partial x$ at the outer surface of the surgeon's glove. After differentiation and integration, one obtains for the amount diffused per unit area

$$Q(t) = \frac{C_o Dt}{d+h} + \frac{2C_o(d+h)^2}{\pi^3 d}\left[\sum_1^{\infty}\left(\frac{(-1)^n}{n^3}\sin\left(\frac{n\pi d}{d+h}\right)\right)\left(1 - e^{-n^2\pi^2 Dt/(d+h)^2}\right)\right] .$$

In the above, with $h=o$ (no surgeon's gloves), the series disappears (since $\sin n\pi = o$) leaving $Q(t) = C_o Dt/d$, the steady-state situation. Furthermore, for $t=\infty$ the series becomes a constant such that $\frac{dQ}{dt} = \frac{C_o D}{d+h} = -D\frac{\partial C}{\partial x}$ or $\left.\frac{\partial C}{\partial x}\right|_{t=\infty} = -\frac{C_o}{d+h}$, as expected.

In applying the results of this computation to glove box usage, it is assumed that sleeves and attendant ventilation are completely effective in preventing tritium exposure to the upper arms. It may be that this assumption is sufficiently wrong to suggest the discarding of this common practice in favor of the wearing of arm-length (obstetrician's) gloves.

The percentage of HTO diffusing through both pair of gloves relative to that with no surgeon's gloves is shown in Fig. 3. The graph is plotted for d = 0.064cm, h = 0.020cm, $D = 5 \times 10^{-8} cm^2/sec$. We obtained the value for D from the permeability and solubility of H_2O in the gloves[2] and Henry's Law. One has from Fick's Law that the permeability P is given by $P = D \frac{\Delta C}{d}$. But C = kp, where k is the solubility constant of H_2O in rubber and p is the partial pressure of H_2O. One has therefore $D = \frac{Pd}{k\Delta p}$.

The values (NTP) $P = 1 \times 10^{-6} cm^3/sec/cm^2/mm$ thickness and k = $150 cm^3/cm^3$ rubber/atm. pressure were assumed. It should be noted that the selection of these particular values was rather arbitrary in that the value of P, which varies widely for various rubbers, was a typical value found by Barrer[2] and others[3]. Unfortunately, the solubility constant k has received little attention, particularly at low humidities. In this case, the value selected was based on the low pressure data of Lowry and Kohman[4].

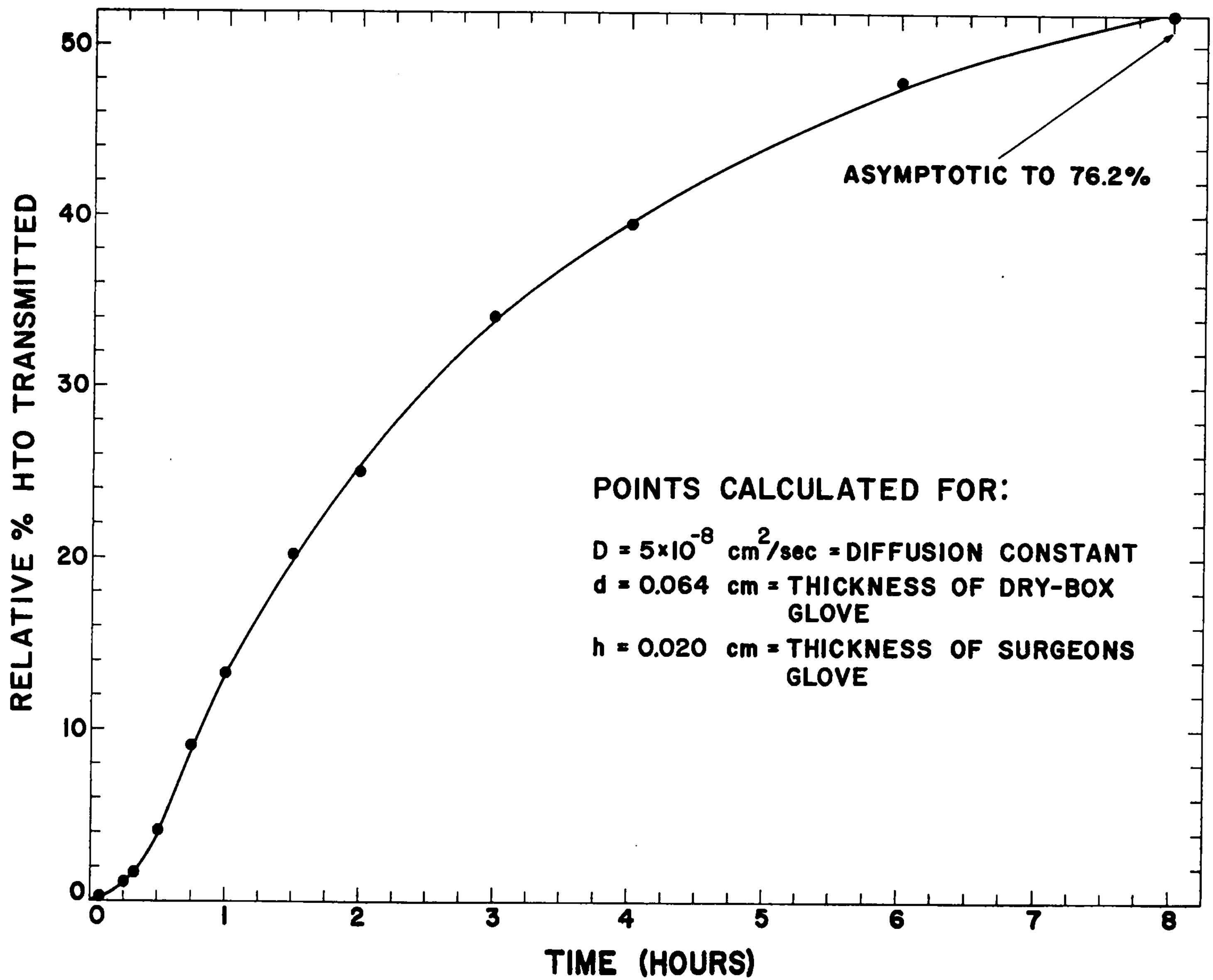


Fig. 3. Relative Permeation of HTO

It is worth mentioning that since D and t appear only as the product Dt in the diffusion equations, an error in D by a given factor will change the time scale on the graph by the inverse factor; the general shape of the curve, however, will not change. Thus, when D is better known, the time scale can be adjusted accordingly. Also note that the curve is asymptotic to 76.2% rather than 100% since even after steady-state conditions occur, the concentration gradient is reduced to $\frac{d}{d+h}$ of its value by using surgeon's gloves.

From the graph, one can choose a reasonable time interval between glove changes. For this purpose, C_o was assumed to be 7.6×10^{-5} cm^3 HTO/cm^3 rubber (corresponding to a concentration of 1 curie per cubic meter in the dry-box gas), and the area was assumed to be that of the area of the surgeon's gloves, or $1500 cm^2$. Thus, for an 8 hour day, the quantity of tritium diffusing out of this area of the dry-box gloves is:

$$Q(t) = \frac{C_o Dt}{d} = \frac{(7.6 \times 10^{-5})(5 \times 10^{-8})(28,800)}{0.064}$$

$$= 1.7 \times 10^{-6} \text{ cm}^3 \text{ HTO/cm}^2 \text{ glove,}$$

or $(1.7 \times 10^{-6})(1500) = 2.56 \times 10^{-3}$ cm^3 HTO/pair of gloves

$$= (2.56 \times 10^{-3})(1.3 \text{ curies/cm}^3 \text{ HTO})$$

$$= 0.0033 \text{ curies/day per pair of gloves.}$$

This quantity can then be compared with the allowable working day intake of tritium of 0.08 millicuries. The latter is based on a maximum permissible body burden of 1 millicurie[5] and a biological half-life of 12 days [6]. Since the allowable intake is a factor of 50 lower than the quantity of tritium diffusing through the dry-box gloves, the surgeon's gloves should be changed every 20 minutes or so.

Admittedly of incidental interest, we can investigate the feasibility of airing out these contaminated surgeon's gloves for subsequent reuse. We follow a procedure by Barrer[7] who estimated the diffusion constant from the efflux of gases from a metal plate. Knowing the diffusion constant D, we can find the time needed for a certain fraction of the initial HTO to diffuse from the glove. Using the relation [7]

$$t = \frac{\pi h^2}{256D}\left(\frac{Q_o}{Q_o - Q}\right)^2$$

we find that the times needed for 50%, 90%, and 99% of the HTO to diffuse out from the gloves are 6.7 min., 3 hours, and 300 hours, respectively. This indicates that it is not feasible to air the surgeon's gloves in hopes of subsequent reuse if we want only a negligible fraction of the initial HTO concentration left.

An additional calculation was made to learn the time in which new dry-box gloves reach steady-state conditions after replacement. The result was that the rate of tritium permeation reaches 90% of that of the steady-state situation within 5 hours after replacement.

In summary we see that the use of surgeon's gloves greatly reduces the HTO radiation hazard to the worker. For values of t up to about ½ hour in Fig. 3 the relative dose of HTO varies essentially with the square of the time. Accordingly if the HTO concentration in the dry-box system is reduced by a factor of 4, then for the same radiation dose one needs to change surgeon's gloves only half as often, i.e., every 40 minutes or so, and vice versa. The identical relationship holds if one works in the dry-box system for only 2 hours per day instead of 8 hours. Thus exposure time and the dry-box HTO concentration are important parameters in choosing a safe working procedure.

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