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## Tritium Protective Clothing

MASTER

T. P. Fuller  
C. E. Easterly

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TRITIUM PROTECTIVE CLOTHING

T. P. Fuller and C. E. Easterly

Submitted by T. P. Fuller to the Graduate School of the University of North Carolina in partial fulfillment of the requirements for the degree of Master of Science in Public Health.

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## GLOSSARY OF SYMBOLS

A Cross-sectional area of the membrane ( $\text{cm}^2$ )

$\alpha$  Pressure constant (cm Hg)

C Concentration of the penetrant ( $\frac{\text{stp cm}^3}{\text{cm}^3}$ )

D Diffusivity of the penetrant in the membrane ( $\frac{\text{cm}^2}{\text{sec}}$ )

J Flow rate ( $\frac{\text{stp cm}^3}{\text{sec}}$ )

P Permeability  $\frac{(\text{stp cm}^3)(\text{cm})}{(\text{cm}^2)(\text{sec})(\text{cm Hg})}$   
Volume of gas at stp ( $\text{stp cm}^3$ ) per second (sec) passing through a cross sectional area of a membrane ( $\text{cm}^2$ ) of thickness (cm) with a specified gas pressure difference across the membrane (cm Hg).

p Pressure (cm Hg)

Q Flow ( $\text{stp cm}^3$ )

stp Standard temperature and pressure

S Solubility of the penetrant in the membrane  $\frac{\text{stp cm}^3}{(\text{cm}^3)(\text{cm Hg})}$

T Temperature

t Time (sec)

V Volume ( $\text{cm}^3$  stp)

$\Delta x$  Membrane thickness (cm)

HTO Tritiated water

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## TRITIUM PROTECTIVE CLOTHING

T. P. Fuller and C. E. Easterly

## ABSTRACT

Occupational exposures to radiation from tritium received at present nuclear facilities and potential exposures at future fusion reactor facilities demonstrate the need for improved protective clothing. Important areas relating to increased protection factors of tritium protective ventilation suits are discussed. These areas include permeation processes of tritium through materials, various tests of film permeability, selection and availability of suit materials, suit designs, and administrative procedures. The phenomenological nature of film permeability calls for more standardized and universal test methods, which would increase the amount of directly useful information on impermeable materials. Improvements in suit designs could be expedited and better communicated to the health physics community by centralizing developmental equipment, manpower, and expertise in the field of tritium protection to one or two authoritative institutions.

## 1. INTRODUCTION

### 1.1 Overview

Protective clothing is a major factor in reducing occupational exposure to radiation at facilities handling high inventories of tritium. Historically, protective ventilation suits have been developed as needed by individual institutions. Materials were chosen from qualitatively acceptable groups of immediately available films. They were picked according to previously known characteristics, principally permeability, fabricability, and cost. Over the past 20 years, suits providing acceptable barriers to tritium have been developed and continuously improved. The acceptability of presently available protective clothing is now being questioned in light of current progress toward the development of fusion reactors and the large tritium inventories they are expected to contain. Therefore, it is desirable to perform an up-to-date assessment of improvements that should be made in protective clothing capabilities.

Tritium protective suits available today are generally considered acceptable in relation to exposure limits and the concentrations encountered at tritium facilities, but this may not be true in the future. Ventilation protective garments currently being used make it possible to work in tritium contaminated atmospheres with concentration levels of 30 mCi/m<sup>3</sup> of HTO at 40 hr/wk without surpassing exposure limits.<sup>1</sup> By controlling the work time with respect to concentrations commonly encountered and permissible exposure levels, these suits provide an adequate means of protection for most situations in which tritium is encountered. With the recent discussion in radiation control organizations about lowering occupational exposure, the adequacy of present

suits is lessened as working times could decrease to unacceptable levels. Potential tritium problems associated with fusion energy provide other important reasons for improving protective clothing. In conceptual fusion facilities tritium inventories may exceed 20 kg with a total activity of  $1.92 \times 10^8$  Ci ( $1g = 9,600$  Ci).<sup>2</sup> Because tritium will be contained in nearly all of the fusion reactor subsystems, high concentrations of tritium are likely to be found in work areas requiring regular hands on attention. The importance of improving protective clothing capabilities stems from:

1. The requirement that fusion power plants have a high degree of operational reliability, necessitating maintenance during operation under potentially adverse conditions;
2. The provision of economically based alternatives to multiple containment schemes; and
3. The necessity of having adequate protection for accident response.<sup>3</sup>

Improvements in protective clothing can be initiated in several different areas. Many of these improvement areas are pointed out in fulfilling the objectives of the present study, which are:

1. To describe important aspects of permeation and polymer film testing relative to the selection of improved suit materials;
2. To communicate information on the availability and practicality of various protective clothing materials;

3. To describe advantages of various suit designs and recent improvements; and
4. To provide a synopsis of research currently underway and indicate some possible future goals in administrative and scientific areas.

### 1.2 Considerations in Tritium Protection

The radioactive isotope of hydrogen known as tritium presents a radiological hazard to workers in many different occupational environments. These include light and heavy-water reactors, breeder reactors, fuel reprocessing plants, tritium production reactors, and in the future, thermonuclear reactors. Each of these systems will have its own radiological hygiene problems concerning tritium. The type of protective clothing will depend upon working conditions along with containment chamber tritium concentrations and activities. The chemical state of tritium is also an important factor in determining the correct protective equipment. Tritium exists as a gas (HT), or in combination with oxygen as a liquid (tritium oxide, HT0), or as a tritiated hydrocarbon. Chemically, it reacts generally the same as hydrogen or water vapor with slightly modified rates. In any chemical form it decays with a half-life of 12.3 years by emitting a beta particle with a maximum energy of 18 keV.<sup>4</sup> These weak betas having a range of 0.005 mm in skin present no external hazard since dead outer layers will attenuate the radiation.

Hazards are associated with the inhalation, or percutaneous absorption, of the oxide or gas which then disperses and exposes internal tissues uniformly through incorporation in the body water. Exposure to

tritium oxide is about four orders of magnitude more hazardous than elemental tritium since the lungs absorb 99.9% of the tritium they encounter as the oxide but only 0.05% of the tritium gas inhaled.<sup>5</sup> The oxide is absorbed at about the same rate through the skin as in the lungs [0.84  $\mu\text{Ci}/(\text{cm}^2\text{-min})$  per  $\mu\text{Ci}/\text{cm}^3$  in air at 30°C and relative humidity of 70%],<sup>6</sup> but the gas is absorbed through the skin at a much lower rate.<sup>7</sup> Problems with tritium protection come from its ability to disperse freely in air and diffuse or penetrate through a wide variety of substances including metal, rubber, and plastic. Tritium oxide is more difficult to contain than HT in that it permeates rubber 100 times faster than HT and 20 times more effectively in cellulosic materials.<sup>8,9</sup> It is practically impossible to assure absolute confinement. Nevertheless, there are some effective methods of tritium control.

Ventilation systems maintaining correct negative pressure are used to decrease tritium concentrations in contaminated areas and prevent contamination of other working environments. Filters utilizing oxidation and reduction reactions are also used to reduce HT and HT0 activities in containment chambers. Protective clothing is probably the most important factor in reducing occupational tritium exposures. Different types of ventilation outfits offer various levels of protection. One protective clothing unit developed in France offers an exceptional protection factor of 16,000 while other suits may only reduce exposure by less than 100, (ref. 10). Together, all the protective devices mentioned above make it possible to work for extended periods of time in areas that would otherwise be uninhabitable. However, in industries with large tritium inventories such as heavy-water fission reactors (where tritium

oxide contributes over 1/3 of the total occupational dose)<sup>11,12</sup> the availability of improved protective measures would be very beneficial.

## 2. PERMEABILITY

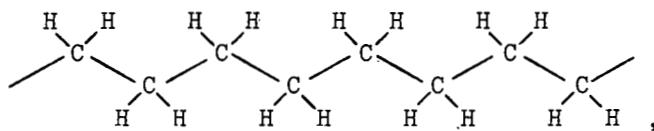
The study and development of less permeable materials is essential to the long-term improvement of protective clothing. In some facilities where tritium is handled, the greatest occupational dose is received via the permeation of tritium oxide through the protective material.<sup>13</sup> Although benefits of research in this area are sometimes outweighed by the costs, further scientific interest may prove to be advantageous especially if a considerably improved material is developed. A general understanding of permeation mechanisms, protective film characteristics, and factors influencing permeability is desirable when selecting and testing films for low permeability.

### 2.1 Membrane Characteristics

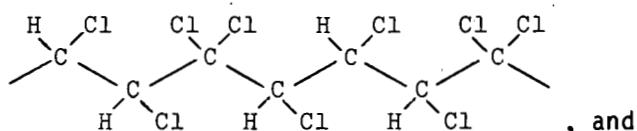
Polymers with low moisture permeability have been found by Morgan<sup>14</sup> to possess some specific molecular characteristics. These are:

1. a saturated or nearly saturated hydrocarbon chain,
2. a minimum of chain branching,
3. a high degree of lateral symmetry,
4. a fair degree of longitudinal symmetry,
5. a very high proportion of relatively small, hydrophobic substituents, and
6. the ability to crosslink between polymer chains.

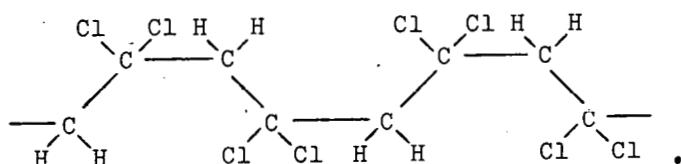
Present technical processes make it possible to create a wide variety of polymers incorporating the above mentioned qualities. The structural characteristics of some common polymeric barriers are:



Polyethylene



Chlorinated neoprene



Vinylidene chloride

Through various methods of production, many different properties decreasing permeability can be incorporated into different polymers. Grafting is the induction of crosslinkage between polymeric chains using chemical and physical methods (including irradiation) to alter molecular structure.<sup>15-18</sup> Polymeric structures with a high capacity for cross linkage and minimal chain branching offer little molecular assistance for diffusing elements. The molecular continuity contributed by grafting inhibits permeation processes in which bonds are formed and the solute concentration is increased in the initial layers of the membrane. Physical changes in the density and cross sectional area can also inhibit Brownian-type diffusion of larger molecules.<sup>19</sup> The most effective membrane materials are essentially nonpolar since hydrophobic materials are generally much less permeable to moisture than hydrophilic polymers. This is due to the increased ability of a polar molecule,

such as water, to form strong polar bonds with and diffuse easily into the initial layer of the membrane surface. The solution of water in the initial membrane molecules also causes physical changes of the polymer such as swelling, which increases diffusion through the expanded lattice that is formed.<sup>20,21</sup> This phenomenon accounts to some extent for the increased permeation rates for the more polar H<sub>2</sub>O molecule than the nonpolar H<sub>2</sub>T gas. Other areas of research showing promise in membrane technology involve ion exchange selectivity<sup>22-24</sup> and the retention of solvents in the protective films.<sup>25</sup>

## 2.2 Permeation Processes

The permeation of substances through polymers is the result of several independent processes. The predominant mechanism of penetration is the molecular streaming of gas or vapor molecules through cracks, capillaries, or amorphous regions in the polymer film. This occurs independently of the type of gas molecule when diameters of holes are less than the mean free paths of the molecules, but are several times greater than the diameters of the gaseous molecules.<sup>25,26</sup> The gas permeability constant for this process K (Knudson flow), defined as the rate of flow per unit area membrane is supplied by the general expression

$$K = B_0 \Delta p/n + K_0 , \quad (1)$$

where  $B_0$  is a geometric factor for the membrane,  $\Delta p$  is the pressure difference of the gas, and  $n$  is the viscosity of the gas.<sup>27,28</sup> This Knudson permeability coefficient decreases with increasing temperature and with the molecular weight of the gas; the coefficient is pressure dependent.

Another mechanism for permeation is diffusion. In this process the permeate creates a boundary layer at the surface of the film and then dissolves into the membrane, moves through under the influence of a concentration gradient, and reevaporates downstream.<sup>20</sup> This transport process consists of the following steps:<sup>20,29</sup>

1. movement in and through the boundary layer,
2. sorption into the membrane,
3. diffusion through the membrane,
4. desorption out of the membrane, and
5. movement out through the boundary layer.

Each of these processes present a different resistance according to the characteristics of the penetrating substance and the phenomenological parameters of the membrane system. Some of the factors influencing permeability are demonstrated in the schematic cross section of a membrane in Fig. 1. In this diagram the physical variables of membrane thickness ( $\Delta x$ ), pressure differences on either side of the film ( $P_1 - P_2$ ), and concentration gradient ( $C_1 - C_2$ ) are represented.

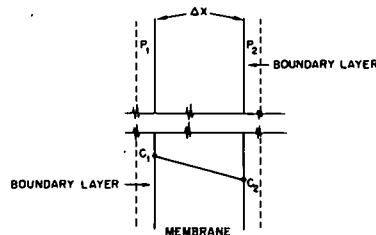


Fig. 1. Homogeneous membrane cross section.

The diffusion of a substance across a membrane can be described by Fick's second law.

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

This equation states that the rate of change in the concentration (C) with respect to time (t) is equal to the diffusivity times the extrapolated rate of change in concentration over a thickness (x). When C remains constant in time equation 2 reduces to Fick's first law:

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

where J is the flux. The concentration gradient is the difference in concentration over the membrane and is represented by

$$\frac{\Delta C}{\Delta x} = \frac{C_1 - C_2}{\Delta x} . \quad (4)$$

Combining this with Fick's first law, (equation 3), gives

$$J = \frac{D(C_1 - C_2)}{\Delta x} . \quad (5)$$

Equation 5 demonstrates the influence of the concentration gradient on permeation. By Henry's law

$$C_1 = SP_1 \text{ and } C_2 = SP_2 . \quad (6)$$

Thus, equation 5 can be rewritten in terms of the difference in pressures since the solubility constant (S) remains the same.

Hence,

$$J = \frac{Q}{tA} = \frac{DS(P_1 - P_2)}{\Delta x} \quad (7)$$

Since permeability is equal to DS,

$$P = \frac{Q}{t} \left[ \frac{\Delta x}{A(P_1 - P_2)} \right] \quad (8)$$

If the downstream volume and temperature are known, the amount of gas passing through the membrane ( $Q/t$ ) can be calculated in standard units of ml/s from the slope of the downstream pressure versus time curve ( $\partial p_2 / \partial t$ ). Using the ideal gas law to relate this information it is possible to calculate permeability,

$$P = (V) \left( \frac{T_0}{T} \right) \left( \frac{1}{\alpha} \right) \left( \frac{\partial p_2}{\partial t} \right) \left( \frac{\Delta x}{A(p_1 - p_2)} \right) \quad (9)$$

In order to see how the individual variables influence permeability, some of these will be discussed in detail below.

The temperature dependence of permeability over reasonable temperature ranges can be given by combined Arrhenius type equations for diffusion and solubility:<sup>9,17,19,20,30,31</sup>

$$S = S_0 \exp \left( \frac{-H_s}{RT} \right), \quad (10)$$

$$D = D_0 \exp \left( \frac{-E_D}{RT} \right), \quad (11)$$

where  $H_s$  is the heat of solution and  $E_D$  is the activation energy for the diffusion process. Combining these two equations with respect to permeability forms a similar relationship<sup>9,31-34</sup>

$$P = P_0 \exp \left( \frac{-E_p}{RT} \right), \quad (12)$$

from

$$E_p = H_s + E_D. \quad (13)$$

Solubility is much less affected by temperature changes than diffusivity, which follows temperature changes to a varying extent depending on the activation energy,  $E_D(T)$ .

Time must be considered in determinations of permeability. When the permeation process being tested approaches steady state conditions the terms for the amount diffusing in time become

$$Qt = \frac{DC_1(\Delta t)}{\Delta x}. \quad (14)$$

Here  $\Delta t$  represents the time ( $t$ ) it takes for a substance to permeate through the film minus the time it takes the system to reach a steady state, time lag ( $L$ ). Plotting  $Q$  versus  $\Delta t$  gives an intercept of  $L(0A)$  on the  $t$  axis shown in Fig. 2. Using this value of  $L$  in the extrapolated relationship

$$D = \frac{(\Delta x)^2}{6L}, \quad (15)$$

it is possible to calculate  $D$  for permeation expressions.<sup>8</sup>

Variations in humidity which affect diffusivity are not completely understood, but they must be considered when testing materials for permeability. Since diffusivity is concentration dependent, it will be

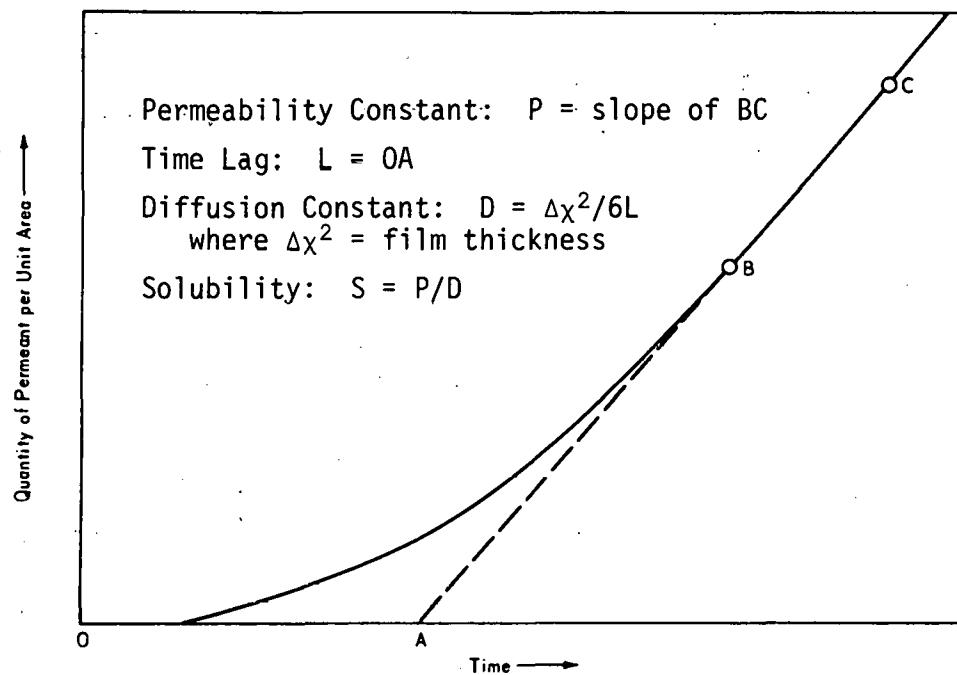


Fig. 2. Typical experiment using the dynamic test method.

affected by changes in humidity. Work done by Hosler showed that when a film specimen is introduced to a new ambient humidity, surface concentrations were altered to new unpredictable equilibrium values.<sup>35</sup> The divergence in data obtained at different humidities is explained by the stress dependence of diffusivity. Different materials respond to changes in concentrations differently. Physical alterations in the polymer's molecular structure due to swelling caused by sorption may lead to increased or decreased permeation depending on that specific system. Information on diffusivity of specimens under external tension or compression would be helpful in determining the effects of stress caused by humidity on permeability.

The formulas presented above are general descriptions of permeation processes independent of experimental methods. Results can be noted in a number of easily convertible units. It must be remembered, however, that permeability is contingent on the phenomenological terms of the system. Variations in the test conditions alter the calculated permeability coefficients irrespective of the general formulas. Changes in thickness, for example, can only be accurately compensated for by the initial equations when the film is completely homogeneous at all thicknesses and the bulk concentration of the permeate remains relatively the same at all points across the film. Otherwise complex calculations must be incorporated into the general permeability formulas to account for any variations.<sup>36-40</sup> Some variables like temperature can be accounted for simply as functions represented in special equations. Alterations in other variables such as humidity cannot presently be accounted for at all since their affects on permeability are incompletely understood. In the past, much of the polymer testing community has hindered optimal understanding within the field by not noting or accounting for the above mentioned variables in presentations of permeability data. In order to best communicate quantitative information about tritium permeation, standardized test methods specifying pertinent variables are required.

### 3. PERMEABILITY MEASUREMENT

Permeability measurements utilize two general methods of testing, one depending on transmission and the other on sorption-desorption principles.<sup>31</sup> Transmission methods involve a concentration gradient across a polymer film separating two sections of a chamber. Various techniques can then supply information on the rate of transmission. Sorption-desorption methods enable calculations of diffusion and solubility coefficients by measuring the uptake and withdrawal of the solute in the film.

#### 3.1 Transmission Methods

Transmission methods for measuring permeation rates through films employ various types of tests. The most common methods involve measurements of pressure, volume, and concentration related variables.

Volumetric test methods depend on alterations in volume caused by penetration of a gas or liquid at constant pressure.<sup>31,41</sup> One volume related permeability test involves the measurement of mercury movement representing a volume change due to transmitted solute in a cell manometer under vacuum.<sup>42,43</sup> A schematic diagram of the gas transmission cell developed into American Society for Testing and Materials (ASTM) method D1454-63 is provided in Fig. 3.

Manometric methods measure permeation by the transmission of gases and condensable vapors indicated by variations in pressure.<sup>44,45</sup> In this test system, measurement is based on increases in pressure on the low-pressure side of the film in a constant volume. A typical pressure oriented permeability apparatus is depicted schematically in Fig. 4.

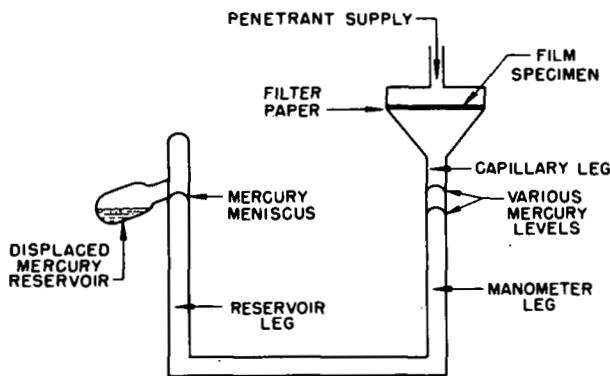


Fig. 3. Schematic for a gas-transmission cell.

The penetrant is supplied to one side of the film, and changes in pressure are measured on the other side using a manometer. Problems in permeability determination using this type of system are associated with the different changes in partial pressure on the low side of the film. They must be accounted for by calibration of the effective volume of the receiving system or in permeability calculations.

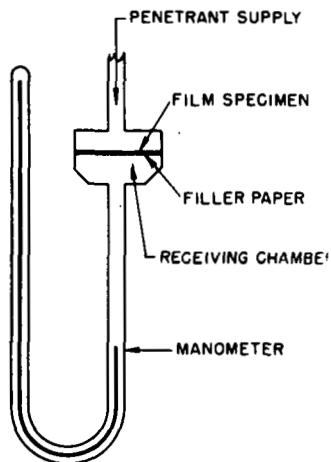


Fig. 4. Schematic of a pressure oriented permeability apparatus.

The concentration associated methods of permeation testing depend on the increase in concentration of the solute in an isolated receiving section of a test chamber. This is a relatively simple and accurate test method which makes permeation measurement possible under a wide variety of test conditions. The most popular permeability testing technique is known as the cup method.<sup>46-48</sup> This test commonly utilizes an assembly that measures weight gain or loss in corresponding test chambers but can involve a number of variations in procedure. The setup consists of two chambers separated by a membrane as shown in Fig. 5. In one cell the solute is introduced under various conditions of pressure, concentration, and temperature. In the other cell there is some way to measure the amount of solute coming through. This is commonly done by measuring weight changes of an absorbing desiccant. As designated by ASTM - E96 (ref. 49), the water-vapor transmission rate *WVT* can be calculated in units of  $\text{g}/\text{m}^2\text{-24 hr}$  from  $WVT = (g \times 24)/(T \times A)$ . Using this relationship it is possible to calculate a relative unit of permeance in metric perms from the following formula:

$$P = \frac{WVT}{\Delta P} = \frac{WVT}{S(R_1 - R_2)} ,$$

$\Delta P$  = vapor pressure difference, mm Hg;

$S$  = saturation vapor pressure at test temperature, mm Hg;

$R_1$  = relative humidity at source;

$R_2$  = relative humidity at sink.

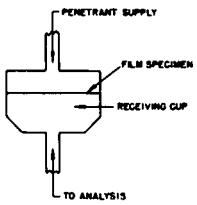


Fig. 5. Detail of vacuum permeability cell.

Dynamic measurement methods using thermal conductivity, chemical analysis, or even radioactive tracers give highly specific quantitative information and make it possible to calculate the permeability constant, diffusion constant, and solubility constant from one experiment. A plot of permeation against time before equilibrium was reached (Fig. 2) makes it possible to calculate a diffusion constant (Eq. 15). The permeability can be determined from the slope of the straight line at steady state conditions. With these two factors known solubility can be found from the relationship  $S = P/D$ . One method devised by Symonds<sup>47</sup> uses radiometric analysis and is especially applicable to measurement of tritium permeation. The system is set up basically the same way as the Payne Cup<sup>46</sup> except that an ionization chamber measures the amount of material solute permeating as the total number of ionizing events occurring within a measured volume of gas. Using the extrapolated time lag technique, solubility and diffusion constants can be calculated to give permeability rates in  $\mu\text{Ci}/\text{ml}$ . If other variables are defined, these units can then be converted to units of  $\text{cm}^3$  of gas at stp penetrating membrane per unit area, ( $\text{cm}^3$ ), per sec for a film thickness (cm) at a gas pressure difference in cm Hg,

$$P = \frac{(\text{stp cm}^3)(\text{cm})}{(\text{cm}^2)(\text{sec})(\text{cm Hg})} .$$

The main advantages of the cup method of permeation measurement stem from the system's ability to alter test conditions and simulate working environments. Permeation of suit material can be measured under various conditions of humidity and partial pressure that would be typical of a ventilation suit in use. With respect to the health physics community, radiometric analysis may be the most practical way to measure permeation of tritium through materials. Permeation units of  $\mu\text{Ci}/\text{ml}$  would not only be a more familiar unit to use in making suit material comparisons but would also make approximations of exposures in these suits somewhat more direct.

### 3.2 Sorption-Desorption Methods

The sorption-desorption methods of permeation measurement are commonly used where solubility and diffusion coefficients are dependent on concentration and large amounts of vapor are sorbed in the system. Experimental data are obtained by suddenly changing the conditions of a polymer's surroundings by varying the activity of the penetrant and determining the amount of penetrant sorbed or desorbed. Solubility is related to the amount of solute picked up by the film immersed in a liquid or suspended in a vapor. Diffusivity represents the rate that vapor is absorbed.

Several test procedures can be used to measure the amount of solute sorbed or desorbed in a film sample. In the weighting bottle method<sup>50-52</sup> the film sample is immersed in liquid or vapor and removed periodically

to be put in a capped bottle and weighed. This process is then repeated periodically until an equilibrium sorption is obtained. Other methods of sorption-desorption testing involve dielectric measurements,<sup>53</sup> helical springs,<sup>54</sup> changes in volume or length,<sup>55</sup> or electrical balances.<sup>56</sup> The main advantage of sorption-desorption measurement techniques is the ability to accurately determine permeability constants for heterogenous materials where the existence of microvoids, fillers, or clusters of sorbed vapor make precise transmission method calculations difficult.

### 3.3 Selection of Test Methods

When selecting materials for protective clothing against tritium or tritium oxide, permeability should be the primary consideration. For this reason a constant unit representing permeability would be desirable in making comparisons of those qualities. Since permeability is a phenomenological coefficient dependent on test conditions, standardized testing is the only method to obtain comparable units. The problem with polymer testing in the past has been the general inability to accurately compare permeability data from different reports. This is because experiments are not commonly done under standardized test conditions, thus resulting in data that cannot be directly compared.

It has been shown that the effect of humidity on permeability is not completely understood and therefore cannot be precisely accounted for in permeability calculations. Variations in other factors such as film thickness, density, temperature, even though understood, introduce complexities into permeability formulas and make comparison difficult. Experimenters often exclude variables and present only relative data.

This limits the usefulness of the data when fully quantitative information could be made available. In an effort to dramatize the problems caused by the lack of standardized testing and reporting, Table 1 is presented. It illustrates the results of some permeability experiments and the ambiguous nature of the permeability constants reported.

Many technologists associate standardization with a bureaucratic system that limits creativity and burdens them with paperwork. Actually, the opposite of this can be true. The goal of standardization is to provide a common language and remove technical barriers to scientific communication. This would reduce confusion and disagreement in data interpretation, speed up distribution of important findings, and decrease the possibility of duplicating experiments. In general, initiating routine procedures increases efficiency by eliminating experimental repetition thus freeing energies for other areas of research.<sup>61</sup>

According to Stanett,<sup>62</sup> several criteria must be considered when determining appropriate standardized test methods. The criteria should be

1. extendable,
2. consistent,
3. unambiguous,
4. convenient, and
5. independent of language.

Usually a standard will be formed with a compromising combination of these. Satisfactory tests are those in which surrounding variables have been conditioned or regulated. For protective suit materials against tritium, test environments should simulate suit pressures, tritium

Table 1. Comparison of permeability coefficients and related variables for H<sub>2</sub>O permeation in polyvinylchloride and polyethylene reported by six different experimenters

Permeability constant (cm <sup>3</sup> )(cm) (cm <sup>2</sup> )(sec)(cm Hg)	Thickness (cm)	Temperature (°C)	Pressure difference, DP (cm Hg)	Relative percent humidity		Chamber volume (liter)	Reference
				Cell 1	Cell 2		
<u>Polyvinylchloride</u>							
1.6 x 10 <sup>-10</sup>	0.019	23°		100		1.5	57
8.1 <sup>a</sup>							58
7.0 x 10 <sup>-7</sup>	0.03	20°	1			13	60
4.0 x 10 <sup>-7</sup>	0.05	20°	1			13	60
14.7 x 10 <sup>-8</sup>							27
3.1 x 10 <sup>-9</sup>	(3-8) x 10 <sup>-3</sup>	23°	2.1	100	0		59
<u>Polyethylene</u>							
2.3 x 10 <sup>-12</sup>	0.005	23°		100	<i>b</i>	1.5	57
5.8 <sup>a</sup>							58
0.74 x 10 <sup>-8</sup>							25
(1.1-1.8) x 10 <sup>-8</sup>	(3-8) x 10 <sup>-3</sup>	23°	2.1	100	<i>c</i>		59
9.7 x 10 <sup>-7</sup>	0.00523	39°	5.3	100	1		14

<sup>a</sup>Measurements are reported in relative units.

<sup>b</sup>Relative humidity varied over the experiment from approximately 0 to about 10%.

concentrations, and other working conditions as closely as possible. This would provide relevant information for the selection of the appropriate materials. In order to insure that developed standards are useful to the development of a field, a complete understanding of the significant characteristics and properties of the material is important. In this case, permeability is the primary concern, so tests should provide specific information on those qualities through a rational system of classification and testing. Secondary factors of less importance (strength, fabricability, etc.) can then be addressed.

## 4. SUIT MATERIALS

With the advent of industries in which the handling of tritium introduced exposure problems, came the need for clothing materials to protect employees. Initially, materials were chosen from groups which had been developed for other purposes and had specifically favorable qualities relative to protection from tritium. These were

1. low permeability,
2. drapability and flexibility,
3. light weight,
4. strength,
5. fabricability,
6. nonallergenic, and
7. availability in quantity at low cost.<sup>63</sup>

In 1938 De Boer studied various celluloid materials for their hydrogen diffusion properties.<sup>64</sup> He found them to be ten times less permeable to hydrogen than vulcanized rubber. Other materials were also tested, but their use was initially inhibited by the further development of an already existing material which adequately provided desired qualities, this was polyvinylchloride (PVC).

In the early 1950's PVC was chosen as the material to be used in tritium protection for two reasons: Firstly, because of its advanced stage of development compared to other polymers and secondly, because of the working experience previously accrued by the scientific and industrial communities. Reference to polyvinylchloride was first made in 1872 by Baumann.<sup>65</sup> He discovered a white powder formed by the action of sunlight

on vinyl chloride in a sealed tube that was extremely resistant to a variety of solvents. From that time significant advances were made in the field of polymer science until 1928 when it was discovered that copolymerization of vinyl chloride with vinyl acetate improved handling characteristics.<sup>66-68</sup> This advance allowed an increase in the practical applications of polyvinylchloride and led to its increased usage and understanding. Combining the inherent properties of low flammability, electrical resistance, low permeability, chemical resistance, and the ability to be processed into a rubbery product<sup>69</sup> with PVC's ability to form relatively light-weight suits that were tear resistant, comfortable, and had little permeation through seams, made the compound a popular product in many industries besides being used for protective clothing. Applications of PVC have been made in almost every product area imaginable with a predicted growth rate of 12% a year.<sup>70</sup> These include packaging materials, piping, automobile parts, appliances, clothing, hoses, credit cards, phonograph records, and innumerable industrial uses.<sup>71,72</sup> As the demand for the product increased, a cyclic pattern of decreased cost (caused by more efficient production methods) followed by increased demand arose. This made PVC more desirable than other materials not only because of its physical characteristics but because it was in a price range that no other product could compete with.

In 1953 P. W. Morgan compiled a paper discussing specific characteristics of structure and moisture permeability of film-forming polymers.<sup>14</sup> He included test data for numerous materials including acetates, Saran, and polyvinylchloride. Although his results demonstrated that other materials, like Saran, were much less permeable than PVC, they were not

developed into clothing because of deficiencies in other respects. Either the polymers were difficult and expensive to produce or they could not be made into comfortable clothing materials because of their allergenic or physical properties. Polyvinylchloride satisfied these other criteria, thus offsetting the drawback of higher permeability. Thus, it became more cost effective to develop PVC further than to expend research to correct problems in other polymers. Any advances made in the properties of PVC would be more rewarding because of the number of its potential uses as compared to other materials. Some materials that had lower permeability than PVC were used for products that could take advantage of physical characteristics. Neoprene rubber, for example, made a poor suit material in the early years because of its cost and weight, but its physical characteristics and relative tritium gas permeability of 0.545 compared to PVC's 1.16 make it an excellent protective glove material.

In 1960 Symonds tested various films which had previously indicated low permeability.<sup>25</sup> He found that of the films tested PVC was the most permeable. It was about 150 times more permeable to water vapor than Saran and 20 times more permeable than polyethylene. These data are presented in Table 2. Along with these results, he published data on effects of specific factors such as layering of films, temperature, and thickness. One important test (Table 3) demonstrated the influence of plasticizers on film permeability. These data indicate that as the concentration of additives, plasticizers, and fillers increases, the permeability of PVC films to water vapor increases. This alteration of permeability emphasizes the importance of film testing on the materials developed for plastic suits rather than the basic film polymers as the permeation characteristics vary significantly.

Table 2. Film permeability to water vapor<sup>a</sup>

Film	Permeability Constant, (cm <sup>3</sup> )(cm) (cm <sup>2</sup> )(sec)(cm Hg)
Aluminum-coated Mylar	<0.1
Saran (vinyl chloride/vinylidene chloride copolymer)	<0.1
Teflon 100X (TFE/HFP copolymer)	0.2
Polyethylene	0.74
Mylar (polyester)	2.2
Polyvinylchloride	14.7

<sup>a</sup>From (ref. 25).

Symonds also published information on the effects of absorbed water in films on the permeation of tritiated water vapor using a copolymer of vinyl chloride and butyl rubber. These data are shown in Table 4. Small amounts of moisture in the film force passages open causing a measurable increase in vapor transmission through diffusion processes. However, large amounts of moisture in the film increased the solubility of tritiated water vapor thus reducing its diffusion and permeation.<sup>27,73</sup> When the film is saturated with water, diffusion paths and microvoids are filled. Penetrating water must expend energy in exchange processes, thus slowing its progress through the film. Symonds also documented the retention and decontamination of tritium in plastic films.<sup>25</sup>

Other important work appeared in the 1960's. In 1962 more information about tritium permeation in neoprene and PVC was provided by Hughes.<sup>32</sup> Some of his results demonstrated that permeation increases with temperature and that the beta particle emitted from permeating tritium has an immeasurable effect on film permeability. Aside from these findings his paper provides a good description of permeation processes. Additional in depth

Table 3. Permeability of PVC films<sup>a</sup>

Percent PVC	Percent ash	Percent plasticizer <sup>b</sup>	Permeability constant, (cm <sup>3</sup> )(cm) (cm <sup>2</sup> )(sec)(cm Hg)	Diffusion constant, cm <sup>2</sup> /sec
<b>A. Film source: Snyder Company (unfilled)</b>				
74.3	0.24	25.7	14.7; 14.9 x 10 <sup>-8</sup>	8.4; 9.7 x 10 <sup>-8</sup>
71.2	0.02	28.6	14.4; 15.1; 15.3 x 10 <sup>-8</sup>	17.2; 24.8; 21.1 x 10 <sup>-8</sup>
70.3	0.36	29.8	16.1; 18.5 x 10 <sup>-8</sup>	16.0; 13.8 x 10 <sup>-8</sup>
<b>B. Film source: B. F. Goodrich Company (filled)</b>				
80.7	9.7	9.6	4.0 x 10 <sup>-8</sup>	6.8 x 10 <sup>-8</sup>
63.4	1.2	35.4	15.1 x 10 <sup>-8</sup>	8.5 x 10 <sup>-8</sup>
62.9	2.1	35.0	16.4 x 10 <sup>-8</sup>	9.8 x 10 <sup>-8</sup>

<sup>a</sup>From (ref. 25).<sup>b</sup>Percent plasticizer = 100 - (percent PVC + percent ash), (not all compositions reported equal 100%).

Table 4. Effect of absorbed water on film permeability<sup>a</sup>

Prior Film Treatment	Permeability constant, (cm <sup>3</sup> )(cm) (cm <sup>2</sup> )(sec)(cm Hg)	Diffusion constant, cm <sup>2</sup> /sec	Solubility, (S = $\frac{P}{D}$ )
Immersed in water 120 hr at 23°C	$8.6 \times 10^{-8}$	$0.73 \times 10^{-8}$	11.0
Film tested after initial test with ordinary water	$14.4 \times 10^{-8}$	$1.3 \times 10^{-8}$	10.9
Conditioned in lab at 60° relative humidity at 23°C	$14.3 \times 10^{-8}$	$1.83 \times 10^{-8}$	7.9
Stored over "Drierite" for six days	$13.6 \times 10^{-8}$	$1.66 \times 10^{-8}$	8.2

<sup>a</sup>From (ref. 25).

information on relative permeabilities of neoprene, butyl, and natural rubber was presented by Ayer.<sup>37,74</sup> His findings are presented in Table 5.<sup>37,74</sup>

Table 5. Relative permeability for various polymeric films at a normalized thickness and water vapor pressure differential across the membrane<sup>a</sup>

Polymer	P (relative)
Butyl-7	2.1
Butyl-6	2.3
Charco milled neoprene	17.0
Neoprene 3	6.7
Supreme 2	9.4
Charco latex neoprene	8.1
Supreme 4	12.0
Cadillac vinyl	23.0
Surety 5	11.0
Surety latex neoprene	23.0
Pioneer latex neoprene	23.0
Charco Hycar	48.0

<sup>a</sup>From (refs. 37, 74).

A study done by Caire gave information on a large number of films including butyl rubber.<sup>58</sup> He found that for many substances the permeation rates for HT and HT0 are inverted. For example, polyethylene is more permeable to tritium gas than PVC but less permeable to tritiated water vapor. His results are shown in Table 6.<sup>58</sup>

Permeabilities for various materials supplied by Billard are given in Table 7.<sup>60</sup> From his study he found Nylon-Butyl to be the best product for protective clothing considering permeation coefficients of HT and HT0, but he could not promote its use because it is not as easy to obtain good seam bonding. Poor bonding causes seams to be less than airtight resulting in higher internal exposures for the completed protective suit. Billard also includes an informative Appendix containing an extensive mathematical derivation of the diffusion processes.

Table 6. Relative permeability of tritium gas in various materials<sup>a</sup>

Polymer	P (relative)	
	HT	HTO
Perbunan rubber	8.4	9.0
Butyl rubber	1.0	1.0
Leaded rubber	0.45	1.6
Scaphair	0.3	25.0
Polyethylene 30/100	12.6	8.1
Nylon reinforced polyethylene	4.8	18.6
PVC 30/100	2.05	5.8
PVC 24/100	2.7	1.4
Nylon coating	6.4	50.0
Nylon butyl	0.2	0.4
Aluminized neoprene	0.067	2.8

<sup>a</sup>From (ref. 58).Table 7. Permeability coefficients for HT and HTO by Billard<sup>a</sup>

Polymer	Thickness ( $\mu$ )	Permeability constant, ( $\text{cm}^3$ )( $\text{cm}$ ) ( $\text{cm}^2$ )(sec)(cm Hg)	
		HT	HTO
PVC	300	1.3	>70.0
PVC	500	1.1	>40.0
Latex	600	4.4	40.0
Cotton-Neoprene	420	4.5	>4.0
Neoprene	640	1.2	4.3
Crystallized Vinyl	460	0.8	>30.0
Nylon-PVC	370	0.23	>15.0
Butasol	390	0.4	0.8
Nylon-Butyl	460	0.4	0.2
Nylane	85	0.3	1.9
Terphane	250	0.14	4.2
Aluminized Mylar	25	0.002	0.5
Terthane OB2	80	0.16	>2.0
Terthane 6A2	130	0.14	1.7
Terthane 6T2	75	0.10	1.1
Saran-Polyethylene	80	0.1	1.5
Saran-Polyethylene	108	0.06	1.2
Saran	90	0.03	0.7
Saran	50	0.02	0.3

<sup>a</sup>From (ref. 60).

Recent permeation studies have concentrated on materials that have specific potential for use as protective clothing. In 1970 Charamathieu found Saran or a copolymer of Saran and PVC to be the least permeable of the materials shown in Table 8, but neither one had desirable mechanical properties necessary for suit fabrication.<sup>75</sup>

Table 8. Permeability coefficients for HT and HT0 by Charamathieu<sup>a</sup>

Polymer	Thickness ( $\mu$ )	Permeability constant, ( $\text{cm}^3$ )( $\text{cm}$ ) ( $\text{cm}^2$ )(sec)(cm Hg)	
		HT	HT0
Technibutyl	1044.0	0.71	0.36
	826.6		
Neoprene	685.8	0.70	1.1
	748.5		
Polyurethane	251.4	0.57	
PVC-Saran-PVC Laminate	419.14	0.11	1.81
	210.71	0.09	1.2
Saran Polyethylene	88.5	0.03	
Crystallized Vinyl	460.0		15.0
Terthene	80.0		1.1
Saran-PVC Laminate	685.0		0.43
Layered Polyester	1450.0	0.02	
Polyethylene-Saran	885.0		0.42

<sup>a</sup>From (ref. 75).

Other tests done by Guild<sup>76</sup> and Derrick<sup>77</sup> indicate relative permeabilities for materials that may be used for parts of ventilation protective suits other than the fabric itself. Derrick demonstrated that Polytetrafluoroethylene (Teflon) had low permeability and would make an excellent support material if cost were not an important factor. Another

study on useful materials was done by Gaevoi<sup>78</sup> who used an elaborate ionization chamber permeation testing device and found permeability constants for several new materials (Table 9).<sup>78</sup>

Table 9. Material characteristics of HT and HTO in some organic materials<sup>a</sup>

Material	Permeability constant, (Ci)(cm) (cm <sup>2</sup> )(sec)(Ci/liter)	
	T	HTO
Natural rubber	$3.3 \times 10^{-10}$	$2.3 \times 10^{-9}$
80/277 PVC formulation	$1.3 \times 10^{-10}$	$1.3 \times 10^{-8}$
L-7 Nairite latex	$9.3 \times 10^{-11}$	$3.7 \times 10^{-10}$
Polyethylene ( $\rho = 0.92$ g/cm <sup>3</sup> )	$5.3 \times 10^{-11}$	$4.5 \times 10^{-10}$
Teflon-2b	$1.7 \times 10^{-11}$	$1.0 \times 10^{-9}$
Terylene	$5.6 \times 10^{-12}$	$3.8 \times 10^{-10}$
Perfol PK-4	$4.6 \times 10^{-12}$	$4.7 \times 10^{-10}$

<sup>a</sup>From (ref. 78).

Development of new materials has been inhibited by the expense associated with the research involved. The science is too complex to consider all desirable properties of a polymer at the same time, consequently, advances have usually come from improvements on specific characteristics of old materials. Dupont has found it cost-effective to pursue the development of additional impermeable materials.<sup>79</sup> This is because they are a large producer of polymer films, and there are many potential uses for improved materials in the sales market.

One of Dupont's newly developed products called Tyvek has many attractive properties with respect to protective clothing against tritium. Tyvek is a spunbonded olefin product made from high-density polyethylene fibers.<sup>80</sup> Spinnbonding is a process in which fine strands

of fiber are spun and interconnected to form a web of filaments arranged at random.<sup>81-83</sup> The threads are then bonded together with high temperature and pressure at their crossover points to form a continuous film. Tyvek can be manufactured to offer a number of different physical properties including cloth-like characteristics. It is strong and has good resistance to chemicals and age degradation. Tyvek is inert, non-allergenic, and meets the wearing apparel requirements of the Federal Flammable Fabrics Act (CS-19-53). Clear Tyvek can be produced without altering physical properties, so it will also make a good suit hood material. Along with the good physical properties mentioned above, a recent study done by Hageman demonstrated polyethylene-Tyvek to be 7.8 times less permeable to tritiated water vapor than PVC.<sup>84</sup> Since that time polylaminated Tyvek has been fabricated into suits by Durafab. The usefulness of these suits cannot be determined until further development and adequate testing facilities are made available.

Another promising material is a Saran-Tyvek lamination. Saran is known to have low permeability, but other physical drawbacks have limited its usefulness. As a protective clothing material Saran is difficult to make at thicknesses that provide the strength and durability needed for use in positive pressure ventilation suits. Laminating it with Tyvek using heat, pressure, and adhesives gives a material with low permeability and good physical properties. In Hageman's study, Saran-Tyvek laminate films were shown to be 150 times less permeable than PVC.<sup>84</sup> DOW Chemical is presently fabricating enough Saran-Tyvek material to make experimental ventilation suits.<sup>85</sup> Durafab then plans to fabricate initial test-run suits, but again, their adequacy must remain unknown in the near future for lack of suit testing facilities.

A disadvantage of Tyvek is its sensitivity to oil and grease which are absorbed in the material. Hydrocarbon solvents can cause Tyvek to swell and permeate through them.<sup>80</sup> This is a problem because tritium associates easily with grease and oil used in mechanical parts and diffusion pumps. The combination of these properties could cause serious problems with use of Tyvek in protective suits. Other problems with Saran-Tyvek laminates are associated with the seams of fabricated suits. Work is currently being done with ultrasound electronics, heat, and pressure seam forming devices to produce strong seams in which permeability is not altered.<sup>86</sup> Problems in this area are not insurmountable. If other methods for providing acceptable seams fail, the seams can always be reinforced by capping them with an extra layer of material.

There are presently three practical materials available to select from when fabricating a protective suit against tritium; neoprene, polyethylene, and PVC. Neoprene is generally less permeable than PVC but more expensive. It has similar draping properties but is normally heavier than PVC because of carbon filler. Polyethylene is about half as permeable as PVC but four times as expensive.<sup>87</sup> It is also a bit more rigid and hood windows must be made of another material because it can not be produced in a clear form. Newly developed materials such as Tyvek and Saran-Tyvek laminations show excellent potential as suit materials, but until they undergo further testing in fabricated suits their usefulness can not be accurately assessed.

## 5. PROTECTIVE VENTILATION SUITS

### 5.1 Wearing Times

Requirements for wearing protective clothing and respiratory equipment are based on the minimization of total dose from external and internal radiation. When protective apparel against tritium is used, the additional dose received due to the lengthening of the time it takes to complete a work task in a radiation area must be considered. The minimum dose theory proposed by Legg is a method to accurately indicate the correct protective equipment for the best level of protection considering all factors that will influence internal and external exposure.<sup>88</sup> In certain instances the wearing of protective gloves may increase work time 50%.<sup>89</sup> The wearing of ventilation suits and respiratory equipment has also been shown to double work time.<sup>88</sup> Unless the protective gear provides significant protection against tritium, the minimum dose theory is a necessity in controlling exposure. Hopefully, the protection factors provided by ventilation clothing in the future will be high enough so that the importance of lengthened work time is diminished. Presently the extent by which work time is increased is still an important factor in the selection of ventilation suits.

If suits provide adequate protection factors, then general tritium protection guides can be used to indicate the protective equipment that is needed in relation to possible tritium exposures. The Savannah River Plant designates the protective apparel to be worn by determining the contamination potential of various areas and labeling them Clean, Regulated, and Radiation.<sup>90</sup> Apparel requirements range from no protective clothing in clean areas to the use of two piece air-supplied suits

prescribed on predesignated time-concentration-based criteria such as demonstrated in Fig. 6.<sup>91,92</sup> At Chalk River National Laboratories (CRNL), air-supplied masks or hoods are worn whenever working in most areas even when chances of tritium exposures are small, because they are easy to wear and not too restraining. They must be worn when the anticipated daily exposure exceeds 8 (MPC)<sub>air</sub>-hours.<sup>93</sup> Since plastic suits are generally more uncomfortable than regular work clothes and a hindrance to work, they are usually not worn until there is an appreciable risk of wetting the skin with tritiated water. The Chalk River protection guides for HTO are summarized in Table 10.<sup>94</sup> Their predesignated acceptable exposure times are demonstrated in Fig. 7.

## 5.2 Suit Design

Several studies have indicated that the degree of protection provided by protective suits and ventilation systems is determined to a large extent by the physical design. The most complete group of tests was done in 1968 by Billard in which he extensively tested several different types of suits.<sup>10</sup> The systems he tested and their efficiencies are described in Table 11. From this table it can be seen that total protection factors up to 16,000 are possible. The efficiency of this particular suit is a result of an extraordinary exhaust system which is impractical relative to the cost, but it nonetheless demonstrates the possibilities. These results tend to show that two piece suits are less efficient than one piece suits, the reasons for which will be described later in this chapter.

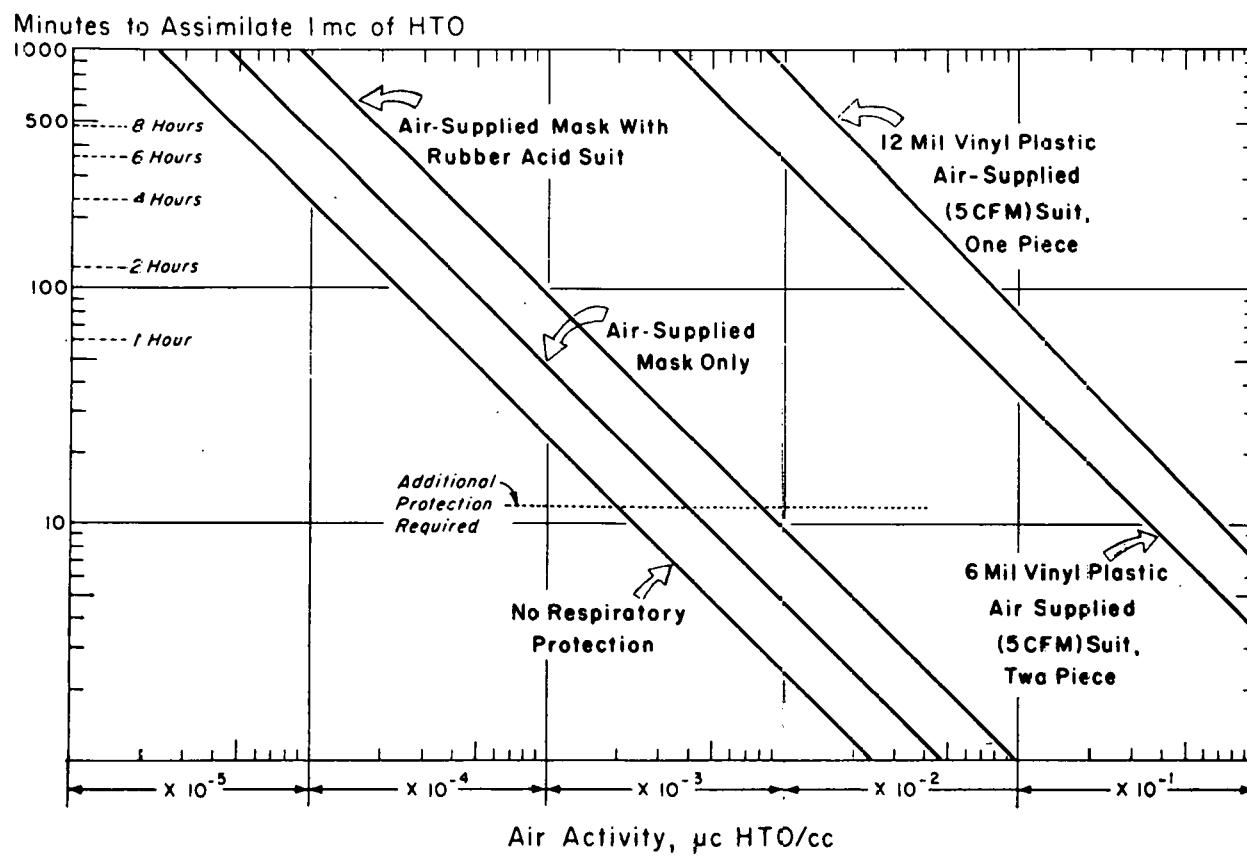


Fig. 6. Nomograph for the determination of working time limits.  
Source: Reference 92.

Table 10. Protection guides for HTO<sup>a</sup>A. For airborne HTO

Anticipated exposure (MPC) <sub>a</sub> -hours per day	Minimum protection
0 to 8	no protection
8 to 30	air-supplied mask or hood
30 to 1500	unventilated plastic suit
1500 to 75,000	ventilated plastic suit
>75,000	assess conditions, estimate intake, and plan exposure carefully

B. When heavy water is present

- Wear an AIR-SUPPLIED MASK or HOOD when working with EXPOSED D<sub>2</sub>O and over OPEN REACTOR HOLES.
- Wear RUBBER or PLASTIC GLOVES when handling WET or MOIST objects.
- Wear PLASTIC SLEEVES when arms might be brushed against WET or MOIST objects.
- Wear a PLASTIC HOOD when there is DRIPPING from overhead.
- Wear a PLASTIC SUIT (including hood) when there is a high probability of BEING WETTED.

<sup>a</sup>From (ref. 94).

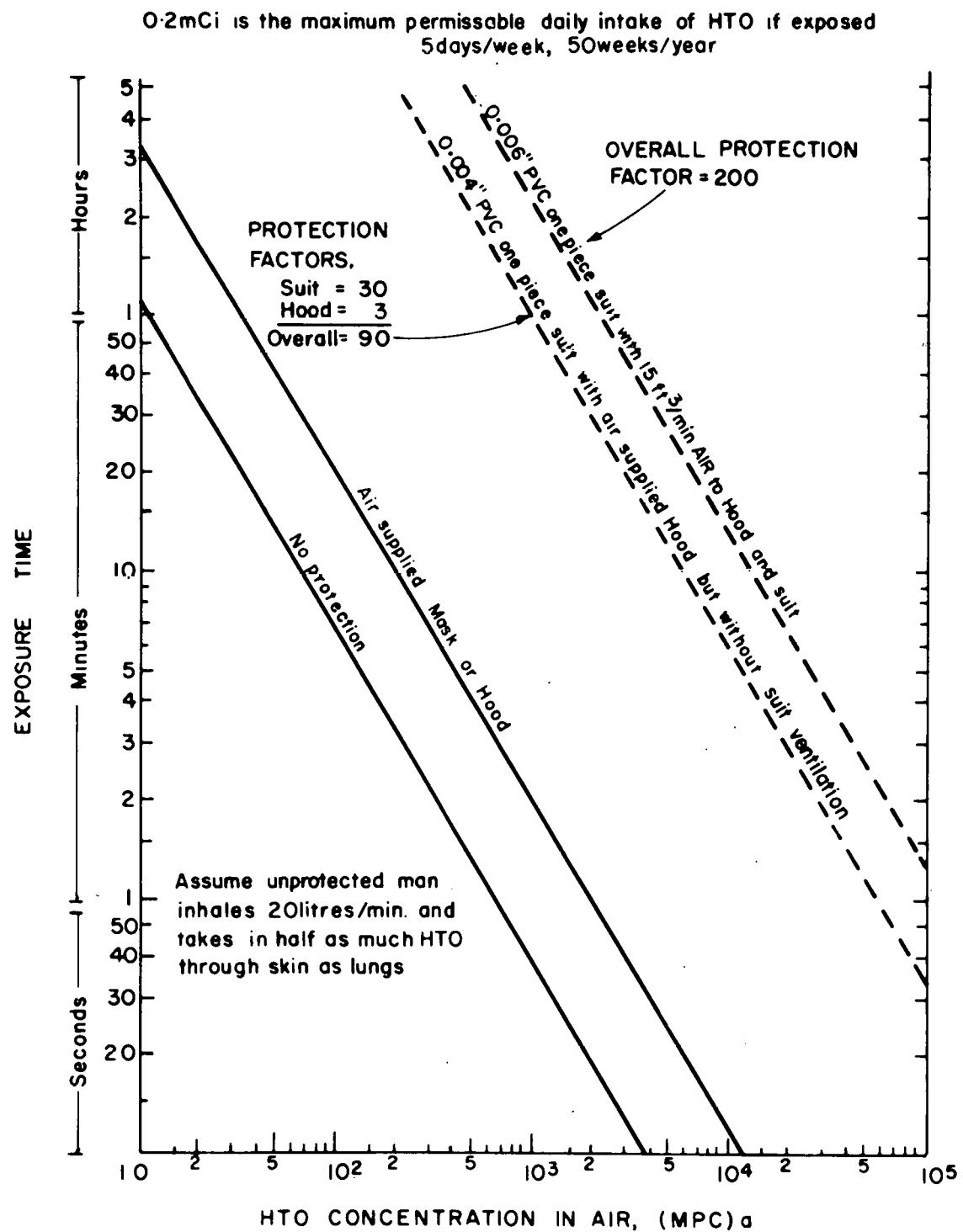


Fig. 7. Time to take in 0.2 mCi of HTO vapor [the 8 hour (MPC)<sub>air</sub> limit]. Source: Reference 93.

Table 11. Ventilation suits tested by Billard<sup>a</sup>

Type	Ventilation	Characteristics	Efficiency
One piece suit, no openings in direct contact with surrounding environment	Distribution of forced air to the hood, arms, and legs	Good air distribution tritium enters mostly by diffusion	3,000
One piece suit, two zipper type closures	Air supplied from the back of suit, poorly distributed	Tritium enters suit by backflow through zippers, holes and diffusion	600
Two piece suit with underalls	Air distribution at the body and in the hood	Poor circulation of air	800
Single piece suit with one zipper type closure	Air supply to hood, no other method of distribution	Tritium entrance by diffusion only, but poor air circulation	4,000
Three piece outfit	Air supply to hood only	Tritium entrance through leaks and diffusion, poor air distribution	1,000
Single piece suit and specially designed airtight helmet attached	Air distribution to hood, hands, and feet	Tritium enters by diffusion only. Expensive	>16,000
Single piece suit with removable airtight helmet	Air supply to hood, hands, and feet	Tritium enters mostly through leaks	150

<sup>a</sup>From (ref. 10).

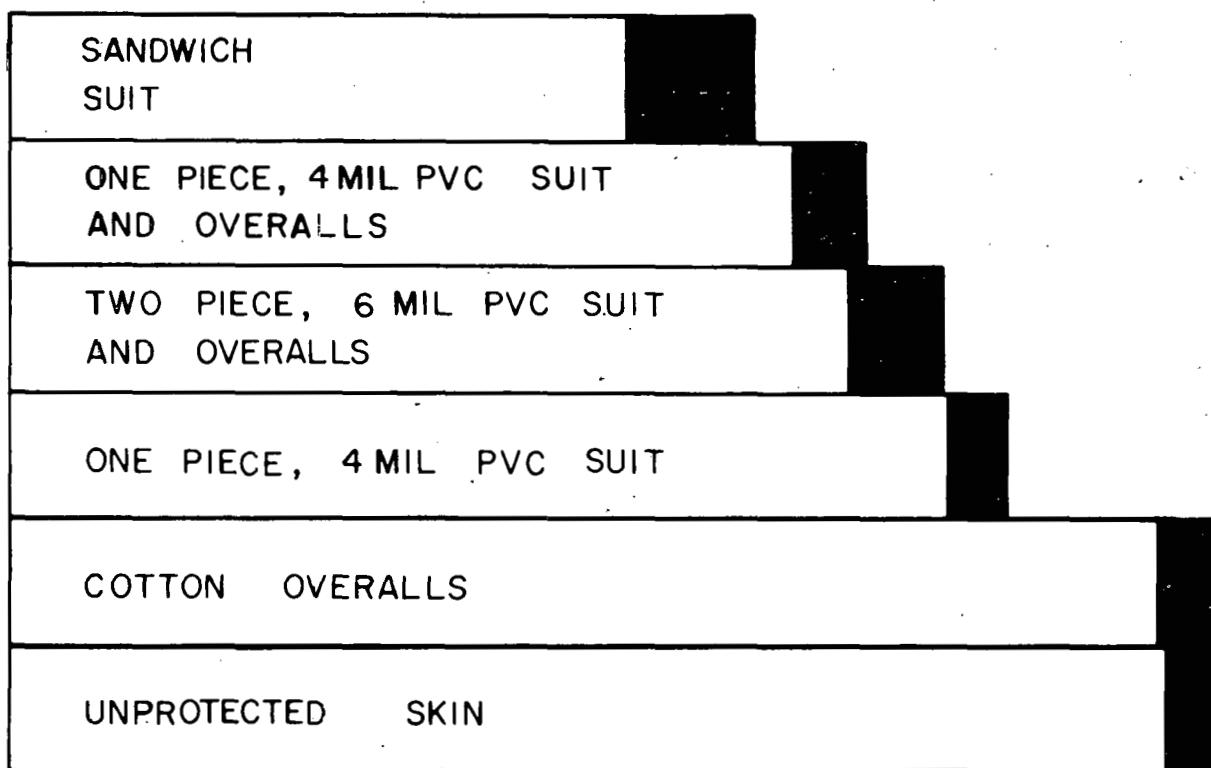
Another study of protective clothing was done by Osborne at CRNL in 1967.<sup>95</sup> The effectiveness of four varieties of protective suits were compared by relative intake rates along with those of cotton coveralls and unprotected skin. His results are shown in Fig. 8. These results were obtained by placing people wearing the suits in exposure chambers and inferring the permeation rates of HTO from intake rates measured through urinalysis. The Sandwich suit, made up from two 4-mil PVC suits with moist cotton overalls between them, provided the best protection. Even so, the application of this type of suit would be impractical because of the complexity involved in dressing or removal and also the restriction of movement and loss of comfort. It also produces different ventilation problems. The one piece 4-mil PVC suit gave nearly the same degree of protection if it is remembered that the sandwich suit used a total of 8 mils of PVC. The data again show that a one piece suit is generally more effective than a thicker two piece suit. The importance of this advantage is somewhat decreased in the final choice of a suit though, because it is more difficult to satisfy other suit criteria, such as ventilation capabilities or comfortability with a one piece suit.

Increased efficiency of single piece suits in comparison to double piece suits was also found in a study by Chassany.<sup>96</sup> He discovered that a two piece PVC suit over cotton overalls and a respirator air supply gave a protection factor of 40. With a one piece suit over cotton overalls and a respirator, there was a protection factor of 120.

Sutra-Fourcade did a study on various types of protective clothing.<sup>97</sup> His findings, which are similar to those noted above, are presented in

## RELATIVE INTAKE RATE

0.001    0.01    0.1    1.0



43

Fig. 8. Summary of effectiveness of protective clothing. Source: Reference 95.

Table 12. Again, it is shown that single piece suits are the best barriers to tritium.

In the past, protective ventilation suits have been developed by individual institutions to satisfy specific criteria of protection by accommodating certain working conditions encountered there. The decision of what protective suit is best is a compromising one. Even though it may provide the least permeation, if the use in the field is limited because of its bulk or other deficiencies, then it may not be the best choice. All qualities must be considered in making the design decision based on the predicted use of the suit. Because of this, suits with common basic features may contain specific characteristics that vary significantly.

The extent by which the penetrant enters the suit through holes, tears, cracks, seams, or even retrodiffusion through exhaust valves profoundly influences the occupational exposure.<sup>26</sup> It follows that the degree of protection is related to the number of parts making up the protective suit<sup>10</sup> since the more suit pieces there are, the more difficult it becomes to obtain an air-tight enclosure. Penetration is also influenced a great amount by the types of closures used in the suit. For example, the use of elastic waistbands has shown to be up to 50 times more effective than drawstrings,<sup>98</sup> and zippers have been found to have poor barrier qualities when compared with other types of closures.<sup>97</sup>

From the data presented it is clear that single piece suits are more efficient than two piece suits; this is mostly due to differences in the number of openings, primarily the one at the waist. The reasons two piece suits may be used in the field instead of single piece ones

Table 12. Protection factors for various types of suits studied by Sutra-Fourcade<sup>a</sup>

Type	Protection factors
Cotton overalls and respirator	2
One piece, 0.1-mm PVC suit and respirator	20-36
Cotton overalls, one piece PVC suit and respirator	120-210
Cotton overalls; two piece, 0.15-mm suit and respirator	40-120
Cotton overalls between 2 one-piece, 0.1-mm PVC suits and respirator	500
Wet cotton overalls between 2 one-piece 0.1-mm PVC suits and respirator	1,900
Two piece air-supplied PVC suit and hood	3,000
One piece PVC suit with two zipper type closures	600
One piece suit with special air flow and removable helmet	800
One piece suit with one zipper type closure	8,000
Three piece suit with adhesive type closures	1,000
One piece suit with specially designed airtight helmet	16,000

<sup>a</sup>Adapted from (ref. 97).

are based on advantages other than protection efficiency. A two piece suit commonly offers a simpler ventilation system than is possible with one piece suits. In principle, the flow of air out from the suit between the shirt and pants limits the circulation of the penetrant against the current. If this is maintained, then the two piece systems' advantages of simple dressing procedures, comfortability and flexibility, and no need for complex closure or exhaust devices make it more practical in use than a single piece suit. Recently, the Savannah River Plant has found a way to reduce the retrodiffusion of the penetrant caused by billowing of the two piece suit by including a porous Tyvek skirt around the waist to limit backflow of air against the current.<sup>99</sup> Another invention that has been quite helpful is a similar containment of the head region with a porous Tyvek skirt around the neck. These devices which solve the problems of backflow along with other superior qualities of two piece suits make the new system very efficient with a protection factor of 10,000.<sup>100</sup>

### 5.3 Ventilation

Air tightness is related to the type of ventilation system provided in the suit. With the problems of backflow in two piece suits solved, they may provide the most efficient practical system available. One piece suit systems are complicated by somewhat restricted movement and the need for exhaust valves which prevent backflow of supplied air at high pressures. Jacques Savornin has developed two exhaust systems that increase the efficiency of one piece suits, but their complexity and costs will probably inhibit their use.<sup>101,102</sup> Most ventilation systems

provide a simple air distribution system through tubes which provide fresh air to the head and extremities. The amount of air a system is capable of supplying through a suit is directly related to the degree of protection afforded.<sup>103</sup> In any event the ventilation system should supply an adequate amount of air to provide comfortable breathing and clear the suit of contaminants in ambient temperatures up to 120°F.<sup>104,105</sup>

Associated with the ventilation system is the noise level in the suit. Los Alamos Scientific Laboratory (LASL) has set the limit at 80 dBA.<sup>106</sup> At Savannah River Plant a plastic suit has been developed with a noise level of 69 dBA at 20 cfm and 73 dBA at 24 cfm of supplied air.<sup>100</sup> This was made possible through two basic alterations in their previous suit designs. First, the air systems "T" connector was replaced with a "Silencer Distributor" (Fig. 9). By eliminating the sharp 90° turn of the "T" and replacing it with the silencer, the air flows through a random path created by a polyester filler, and noise is suppressed prior to the air's entering the hood and extremity sections of the suit. The second improvement was made by replacing the ring header, which supplied air at the top of the hood from a tube with several holes, with a jacket hood plenum filled with open pore polyurethane.<sup>100</sup> This provides a random path for airflow which suppresses and absorbs noise. The floor of the plenum is then made of cardboard, and large holes are used to direct air to the breathing zone. These modifications now make it possible to work for extended periods without the use of headphones or electrical communication systems.

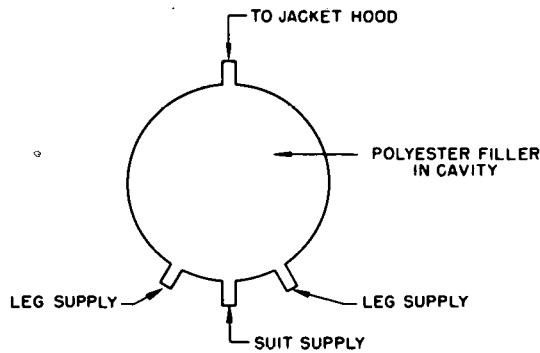


Fig. 9. Silencer distributor.

Not only should the noise levels be palliated for workers subjected to the noise for extended periods of time, but other factors such as heat or confinement should not produce psychological or physical stress either.<sup>105</sup> Suit temperature control can be provided at comfortable levels when an air supply providing 100 psig is coupled to a vortex tube.<sup>92,93,105,107,108</sup> These lightweight devices can be worn to heat or cool the suit to desired levels, making working conditions more comfortable and extending work times. One factor reducing psychological stress and the possibility of accidents is the physical confinement of the suit and the degree of visibility offered by the hood. Lightweight materials make work less exhaustive and increase the level of motor coordination of the worker. Hoods should reduce the field of vision as little as possible and offer plenty of room to accomodate other essential equipment, such as helmet or headphones, when needed.<sup>109</sup>

#### 5.4 Donning and Removal

One important consideration affecting protection that has not been emphasized in the past is the availability of clear concise instructions for donning and removal of equipment. The complete efficiency of a suit cannot be provided unless it is fitted and worn properly. Stephenson has found that technique is probably more important in limiting exposures than the clothing itself.<sup>110</sup> Employees should have extensive training in the properties of the contaminant they are working with, and they should maintain expertise in the selection of protective apparel needed for a specific job.<sup>111</sup> Suit removal is a major factor contributing to the total occupational dose received by workers.<sup>112</sup> Some methods by which exposures received while removing suits may be decreased include showering in suits to wash, then wipe contaminants away, assistance of other workers dressed in protective suits themselves who may cut the workers out of the suits, and the use of pull-tabs for easy release from the suit. In all cases, written dressing and undressing procedures should be provided, and workers should also assume some level of health physics responsibilities for their own safety.<sup>90,92</sup>

#### 5.5 Economics of Suits

The use of some very effective protective ventilation suits may be inhibited by the cost of everyday use by large numbers of employees. The Douglas Point Air-Supplied Suit, for example, has a high degree of protection, but it costs \$500.<sup>105</sup> This may make its common use impractical at facilities using large numbers of them, such as the Savannah River Plant where over 22,000 suits are used each year.<sup>112</sup> In fusion

reactors the large number of workers needing protection from tritium will require that the cost of the protective suits be as low as possible. The Savannah River Plant presently uses disposable suits which cost below \$20. In 1974 the total cost of their program, including all equipment, was \$603,000.<sup>90</sup> One area with potential for reducing costs of clothing is the use of exchangeable inlet and exhaust fittings, another is the laundering of suits. At the Pickering Nuclear Generating Station in Ontario, suits costing about \$100 have the potential for 100 uses being laundered at \$2 per wash.<sup>89</sup> This represents a large savings if laundering procedures do not alter suit efficiency. Tyvek suits offer a new solution to disposal problems. These suits can be incinerated at high temperatures to create CO<sub>2</sub> and H<sub>2</sub>O unlike PVC which converts to acidic chlorine compounds which cause rapid corrosion of metallic parts and may be environmentally unacceptable.<sup>80,106,113</sup> Ease of disposal could be a significant asset when the large number of suits that will be used in a fusion reactor is considered.

### 5.6 Gloves

The use of protective gloves plays a major role in reducing exposure to tritium. At most institutions, double layers of neoprene or polyethylene gloves are worn, and the outer pairs are frequently changed. This is because in working in direct contact with contaminated mechanical equipment, they become saturated with HTO relatively quickly. The protection factors and glove types used at CRNL<sup>93</sup> are illustrated in Table 13. The gloves are often covered with a pair of cotton gloves in order to reduce the chances of tearing the rubber and receiving an unexpected exposure.

Table 13. Protection factors for gloves<sup>a</sup>

Glove type	Estimated protection factor
4-mil PVC	50
7-mil PVC	110
10-mil latex	120
14-mil neoprene	70-340

<sup>a</sup>From (ref. 93).

All of the above factors concerning the design of various protective clothing elements must be kept in mind in developing quality equipment that will be effective, comfortable, and economical. Variations in present suits are due partly to differences in intended use and partly to poor communication of individual advancements made. In order to develop protective clothing capable of satisfying specifications for fusion reactors, more research on effective suit designs is required and better communication of available data between existing test facilities is called for.

## 6. DEVELOPMENT OF PROTECTIVE CLOTHING

## 6.1 Operative Procedure

The design of protective ventilation suits is governed to some extent by specifications documented in reports written at the Los Alamos Scientific Laboratory (LASL). Two publications concerning the design and test procedures of supplied air suits and respirators specify ideal standards for equipment used in conjunction with the Department of Energy (DOE).<sup>106,114</sup> These reports are somewhat general as to the design of the suit, but they do provide specific limitations and requirements which the suits must accommodate for approval. Another guide put out by the International Atomic Energy Agency gives a basic definition of what a protective clothing program should encompass.<sup>111</sup>

In the present system of classification and approval, suits are developed by individual institutions associated with DOE. If the suits satisfy certain operating requirements, they may be sent by DOE to LASL to be tested according to an established procedure.<sup>106</sup> When the suit examination is completed, LASL formulates conclusions and recommendations about the adequacy of the suit and writes a report to be reviewed by an Advisory Committee and by DOE Headquarters for final approval. Improvements may then be made on the existing suit design and, on an informal basis, patterns can be made available to private manufacturers for fabrication.

With respect to the development of protective clothing against tritium, the present program of suit approval contains a few deficiencies. One of the largest problems is due to the operative procedures. Instead

of concentrating available equipment and scientific expertise at a single location to develop specific improvements in protective clothing, the burden of progress has been decentralized and diluted to various user institutions. This method is quite inefficient in that these user institutions normally have neither the manpower nor the facilities to develop or test a large number of improved ventilation suits. It may be thought that decentralization of research efforts would improve progress of suit development, but this has proven not to be the case. Poor communication between researchers, duplication of tests, and the extent of administrative and testing procedures outlined in the LASL report<sup>106</sup> make progress slow, time consuming, and costly. Another problem with the existing situation is that it normally excludes private manufacturing companies from obtaining enough information pertaining to suit development to make any significant advancements or contributions to the field.

## 6.2 Suggestions for Improvement

The faults of the present system could be corrected by providing an institution, which already has tritium testing facilities, with the funding and authority to set up a national testing and development program. An effective organization would provide the following services:

1. develop specifications for protective ventilation suits and the materials used therein,
2. fabricate suits to satisfy their specifications,
3. test developed suits and correct deficiencies,
4. test suits and materials that have been developed by other institutions (governmental or commercial), and

5. provide a continuous source of current information on various types of suits and materials to all interested organizations and enterprises.

The most important aspect of these services is the improvement of communication in the field. Private industries would have easier access to specific criteria of suit design and materials. Being more directly involved, they may find incentives to reduce fabrication costs or improve suit designs in efforts to win bids from major institutions. Facilities using protective suits could still develop suits according to their needs, but they would also have an expert source of information concerning all new advancements made in the field.

The need for a centralized authority which would communicate information has been shown by the fact that improved methods of tritium protection are typically only used at the facilities where they were originated. Frequently, extended time periods transpire before an improvement made at one institution is used at another. Some improvements are not widely used because individual institutions do not have the time or money to thoroughly investigate and incorporate improvements of other facilities into their own programs. The desire to provide the services needed has been made known by a number of groups. The resources currently being distributed to several institutions would be more effectively spent if concentrated in a centralized group.

In the interest of fusion technology, a long-term plan for the development and testing of protective clothing along with a wider range of planning is essential. Increased understanding of its potentials and limitations will aid in establishing the design specifications for areas

where routine work procedures, maintenance, and rescue may be necessary for future fusion power research and production facilities. Early development of protective capabilities should prevent fusion program delays due to inadequacies of devices needed for such operations.<sup>115</sup> More data are needed on the future capabilities of protective clothing with respect to the projected working conditions in fusion reactors. These involve tritium concentrations, available containment, air supply engineering, and number of personnel. More information is needed about these factors before conclusions can be reached about the adequacy of present protective technology.

Emphasis must also be put on new inventions concerned with protection from tritium. Further development of self-contained breathing apparatus and testing of respirator-canister efficiencies involving break-through times of various airborne gas, vapor and particulate contaminants is needed. New methods of contaminated atmosphere ventilation and filtration should also be studied with respect to fusion reactors, as they may reduce the importance of ventilation suits in tritium protection.

### 6.3 Conclusions

The use of protective clothing is a major factor in reducing exposure to radiation in tritium-contaminated areas. With the recent progress made in fusion technology, the call for improved protective devices becomes even more urgent. More coordination between the fields of engineering and health physics must be made in order to accurately assess the need for improved protective equipment at future fusion

facilities. Health physics capabilities concerning protective ventilation suits and development of impermeable materials must be expanded in conjunction with other technological advances. In order to substantially reduce the total occupational dose received at facilities presently handling large amounts of tritium, (1) further work must be done to improve protective clothing capabilities, and (2) better communication of those improvements must be made. Significant improvement in protective clothing is presently possible through the medium of a centralized cooperative program between plastics manufacturers clothing fabricators and evaluation laboratories.

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