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TITLE: A STUDY TO DETERMINE THE EFFECTIVENESS OF PERSONNEL PROTECTIVE EQUIPMENT AGAINST TRITIUM AND TRITIUM/HYDROCARBON MIXTURES

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A STUDY TO DETERMINE
"THE EFFECTIVENESS OF PERSONNEL PROTECTION EQUIPMENT AGAINST TRITIUM AND TRITIUM/HYDROCARBON MIXTURES"*

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I. INTRODUCTION

A. Background

The U.S. Department of Energy (DOE) is concerned whether DOE facilities producing tritium gas (HT) or tritiated compounds are using protective equipment that is "state-of-the-art" and if there is a need to assist these facilities in improving their worker protection.

Gaseous tritium is not significantly absorbed into the body. The exchange of tritium with the hydrogen in body compounds is slow. However, it can be converted to water by oxidation in the atmosphere or by exchange of tritium atoms with the hydrogen in normal water to form tritiated water (HTO). The HTO entering the mouth, lungs, or in direct contact with skin via perspiration is taken up and dispersed throughout the body. Therefore, exposure to high tritium concentrations for any length of time requires skin protection as well as respiratory protection.[1]

B. Problem Statement

DOE requested that the Industrial Hygiene Group of Los Alamos National Laboratory conduct a study to answer the above concerns. The following work statement was developed:

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* Work performed at Los Alamos National Laboratory under the auspices of the US Department of Energy, Contract No. W-7405-ENG-36.

PHASE I: (1) review the literature, (2) survey the tritium protective clothing in use at DOE and other facilities, and (3) if indicated in A and B above, locate laboratory facilities for tritium permeation testing and fabricate/assemble a tritium permeation test system.

PHASE II (contingent on determinations in Phase I): (1) conduct laboratory permeation testing of suit and glove material swatches, (2) develop a suit testing protocol based on results of Phase II Section 1, and (3) conduct full-suit testing if a need is indicated.

II. PHASE I OBJECTIVES

The status of the Phase I objectives is the available literature on tritium permeation testing including a previous review has been completed; all US DOE contractors handling tritium and Canadian facilities at Ontario Hydro and Chalk River, Ontario, have been surveyed to determine the personal protective equipment in current use; a preliminary survey of laboratories where permeation testing could be performed has been conducted; and an evaluation of tritium permeation test methods and apparatus in use, including detectors for the tritium compounds, has been initiated.

III. LITERATURE REVIEW

A. Review Methodology

When initially discussing this study with Los Alamos and Canadian personnel working with tritium, several contacts referred to the document "Tritium Protective Clothing" by T. P. Fuller and C. E. Easterly,[2] which contained a review of the tritium permeation literature up to 1978 and had an extensive reference list. This document and others applicable to this study were obtained at the Los Alamos National Laboratory. Other reference material not easily obtained from libraries or open literature was requested directly from the author or the organization where the investigations were conducted. Literature from 1978 to the present was obtained in this manner.

Tables extracted from individual papers are included in the following section because it was not always possible to recalculate the permeability constants as presented to be compared with other reported units.

The following definition will be used in this report, with exceptions noted in the text:

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

Volume of gas at standard temperature and pressure (stp cm^3) per second (sec) passing through a cross sectional area of a membrane (cm^2) of thickness (cm) with a specified gas pressure difference across the membrane (cm Hg).

The available literature presents data dealing with the permeation of tritium gas (HT) or tritiated water vapor (HTO_v), and liquid HTO_l. Only a limited amount of data was found which discussed the permeation of tritiated organic compounds.

B. Material Tests

Fuller and Easterly's document provides information on permeation processes and calculations. It discusses the two general test methods that are used to determine tritium permeation of materials: (1) the transmission of the penetrant through a polymer film separating two sections of a chamber and (2) a sorption-desorption method that enables calculations of diffusion and solubility coefficients by measuring the uptake and release of the solute by the film. The authors discuss the variability of data caused by the lack of standardized testing and reporting.

This report indicates that in the early 1950s polyvinyl chloride (PVC) was chosen as the material to be used for tritium protection for two reasons: (1) its advanced stage of development compared to other polymers, and (2) the scientific and industrial work experience with it. PVC had many mechanical properties that made it a desirable suit material. These properties are low flammability, low electrical resistance, low permeability, and good chemical resistance. It could also be fabricated into flexible suits which were light weight, tear resistant, comfortable, and had seams sealed against leakage.

Due to discrepancies in Fuller and Easterly's document, their original references were obtained and reviewed again. The data presented below is from these authors and more recent papers. At the end is a summary chart comparing permeabilities of some of the materials as reported by the investigators.

Morgan (1953)[3] discussed the structural characteristics and moisture permeability of polymeric films. His results indicated that materials like Saran were less permeable to moisture than PVC.

Symonds (1960)[4] showed that PVC was about 150 times more permeable to water vapor than Saran, and 20 times more permeable than polyethylene (Table I). He also learned that the permeability of PVC to water vapor increases as the percentage of additives, plasticizers, and fillers are increased (Table II). Symonds' thickness parameter is located in the denominator of the permeability number.

TABLE I

FILM PERMEABILITY BY WATER VAPOR (SYMONDS-1960)

FILM	Permeability $\frac{cm^3}{(cm^2)(cm)(sec)(cm\ Hg)}$
Aluminum-coated "Mylar"	$<0.1 \times 10^{-8}$
"Saran" (vinyl chloride/vinylidene chloride copolymer)	$<0.1 \times 10^{-8}$
"Teflon" 100X (TFE/HFP copolymer)	0.2×10^{-8}
Polyethylene	0.74×10^{-8}
"Mylar" (polyester)	2.2×10^{-8}
Polyvinyl Chloride	14.7×10^{-8}

TABLE II

WATER VAPOR PERMEABILITY OF PVC FILMS BY SYMONDS (1960)

Per cent PVC	Per cent Ash	Per cent Plasticizer[a]	Permeability $\frac{(cm^3)}{(cm^2)(cm)(sec)(cm\ Hg)}$	Diffusion cm^2/sec
A. Film source: Snyder Company (unfilled)				
74.3	0.24	25.7	14.7; 14.9 x 10^{-8}	8.4; 9.7 x 10^{-8}
71.2	0.02	28.6	14.4; 15.1; 15.3 x 10^{-8}	17.2; 24.8; 21.1 x 10
70.3	0.36	29.8	16.1; 18.5 x 10^{-8}	16.0; 13.8 x 10^{-8}
B. Film source: B. F. Goodrich Company (filled)				
80.7	9.7	9.6	4.0 x 10^{-8}	6.8 x 10^{-8}
63.4	1.2	35.4	15.1 x 10^{-8}	8.5 x 10^{-8}
62.9	2.1	35.0	16.4 x 10^{-8}	9.8 x 10^{-8}

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[a] Per cent plasticizers = 100 - (per cent PVC + Per cent ash), (not all compositions reported equal 100 per cent).

Symonds evaluated several thicknesses of Saran and determined that this parameter greatly influenced permeation. (Table III)

TABLE III

PERMEATION DATA ON MULTIPLE LAYERS OF "SARAN" FILM BY SYMONDS (1960)

No. of Layers	Thickness (cm)	Permeability $cc/(sec)(cm^2)(cm)(cm\ Hg)$
1	0.00125	0.22; 0.23 x 10^{-8}
2	0.0025	0.15 x 10^{-8}
4	0.0050	0.14; 0.15 x 10^{-8}
8	0.0100	0.11 x 10^{-8}
1[a]	0.0048	0.14 x 10^{-8}

---

[a] "Saran" film supplied by Snyder Manufacturing Company, Inc.

He then demonstrated that small amounts of moisture in dry film opens up the passages, causing a measurable increase in HTO_v transmission, while in films saturated with water there is a significant reduction in the HTO_v diffusion and permeability rate. (Table IV)

TABLE IV

EFFECT OF ABSORBED WATER ON FILM PERMEABILITY TO HTO_v BY SYMONDS (1960)

Prior Film Treatment	Permeability $\frac{(cm^3)}{(cm^2)(sec)(cm)(cm\ Hg)}$
Immersed in water for 120 hr at 23°C	8.6×10^{-8}
Film tested after initial test with ordinary water	14.4×10^{-8}
Conditioned in lab at 60° relative humidity at 23°C	14.3×10^{-8}
Stored over "Drierite" for 6 days	13.6×10^{-8}

Symonds' results after evaluating PVC, Saran, polyethylene, and Saran-PVC indicated that a laminate of Saran-PVC was the best material to use for suits when permeability, diffusion, and decontamination were considered.

In 1962 Hughes[5] presented HT permeation information for neoprene and PVC. He showed that increased temperature causes an increase in permeability, but the increased β flux emitted from tritium had an immeasurable effect on film permeability.

Caire (1966)[6] studied a large number of films and found for many substances, the permeation rates for HT and HTO are inverted. For example polyethylene 30/100 is more permeable to HT than PVC 30/100 but less permeable to HTO_v. Caire reports permeation rates relative to the permeation of butyl rubber. (Table V)

Katoh (1968)[7] measured the permeation rate of HTO through five PVC films, two PVC decontamination suits, six PVC air-line suits, four polyethylene films, four latex rubbers, and six neoprene rubbers. The initial HTO concentration was 100 µCi/ml. Katoh's results are much lower than those measured by others.

Polymer	Permeability Constant P
PVC	$\sim 2 \times 10^{-10}$
PVC films	$P_{ave} = 1.7 \times 10^{-10}$
PVC decon suits	$P_{ave} = 2.6 \times 10^{-10}$
PVC airline suits	$P_{ave} = 2.9 \times 10^{-10}$
Polyethylene films	0.02×10^{-10}
Latex rubber films	$\sim 1 \times 10^{-10}$
Neoprene rubber films	$\sim 0.4 \times 10^{-10}$

TABLE V

RELATIVE PERMEABILITY OF TRITIUM GAS IN VARIOUS MATERIALS BY CAIRE (1966)

	P (relative)	
Polymer	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

Billard (1968)[8] and Charamathieu (1970)[9] worked together at the Centre d'Etudes Nucleaires de Saclay, France. They determined the tritium permeation data in Tables VI and VII. These data indicate, laminated materials are less permeable than PVC.

Steinmeyer and Braun (1975)[10] measured permeability constants for the permeation of hydrogen, deuterium, and tritium for a number of elastomeric and polymeric materials. The measured values compared quite well with several literature values for hydrogen and deuterium; however, no comparisons were referenced for tritium. Table VIII presents only the tritium measurements. This study indicated that Kapton and then Buna-N rubber are the least permeable to HT.

TABLE VI

PERMEABILITY CONSTANTS FOR HT AND HTO_v BY BILLARD (1968)

Polymer	Thickness (μm)	Permeability $\frac{(cm^3)(cm)}{(cm^2)(sec)(cm\ Hg)} \times 10^{-8}$	
		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 082	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

Gaevoi (1976)[11] designed an ionization chamber apparatus that allowed four specimens films to be examined simultaneously. The challenge concentration of HT and HTO was 0.001 to 1.0 Ci/liter with film thicknesses of 0.0075 to 0.11 cm. The data are presented in Table IX. Curies are used in this permeability number.

Hageman (1978)[12] performed tritium permeation tests for the materials under consideration at that time by Savannah River Plant for suits. The HTO_v that permeated the tested materials was collected and counted each hour for eight 1-hour samples.

He reported that the relative effectiveness of the PE coated Tyvek was seven to eight times better than the PVC to HTO_v permeation. The Saran laminated Tyvek performed 150 times better after 3 hours and 64 times better after 8 hours than did the PVC.

A disadvantage of using Tyvek in tritium handling areas is its ability to easily absorb oil and grease which are frequently tritiated, for example in vacuum systems.

TABLE VII

PERMEABILITY CONSTANTS FOR HT AND HTO$_V$ BY CHARAMATHIEU (1970)

Polymer	Thickness (μm)	Permeability $\frac{(cm^3)(cm)}{(cm^2)(sec)(cm\ Hg)} \times 10^{-8}$	
		HT	HTO
Technibutyl	1044.0	0.71	
Technibutyl	826.6		0.36
Neoprene	685.8	0.70	
Neoprene	748.5		1.1
Polyurethane	251.4	0.57	
PVC-Saran-PVC			
Laminate	419.14	0.11	1.81
Laminate	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
Saran	90.0	0.03	0.16

The following data was taken from Hageman's graphs (Fig. 1, 2, and 3):

PVC:	Slope of 4.93 μCi/hr.
PE-Tyvek:	Slope of 0.63 μCi/hr.
Saran-Tyvek:	Slope of 0.12 μCi/hr.

In 1981, Doughty and West[13] obtained permeation constants for glove materials: neoprene, hypalon, two butyl rubbers, and two laminates EPDM/butyl rubber and hypalon/neoprene (Table X). They used two closed loop circulating gas systems that flowed past opposite sides of a flat sheet of the elastomeric material to be tested and dilute tritiated water as a tracer with a novel method of humidity generation. Humidity was generated by controlled heating of a column of molecular sieves which contain diluted tritiated water.

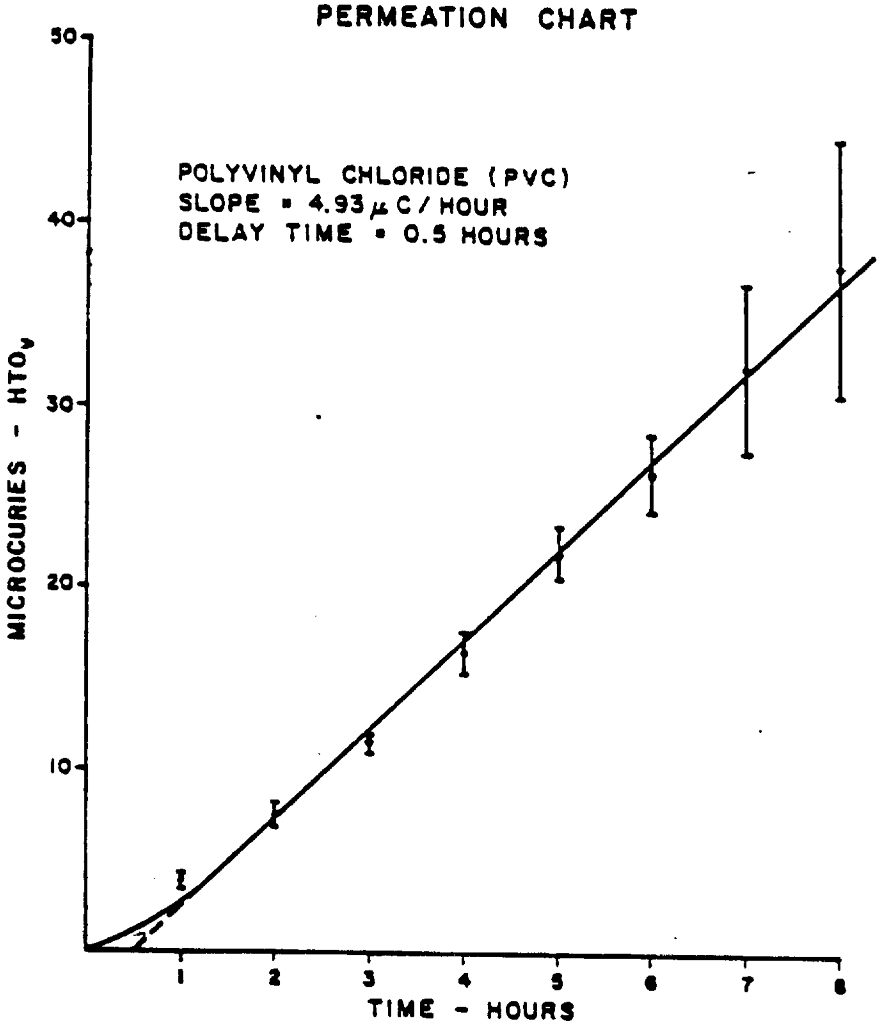

Figure 1 - Hageman Permeation Chart for Polyvinyl Chloride

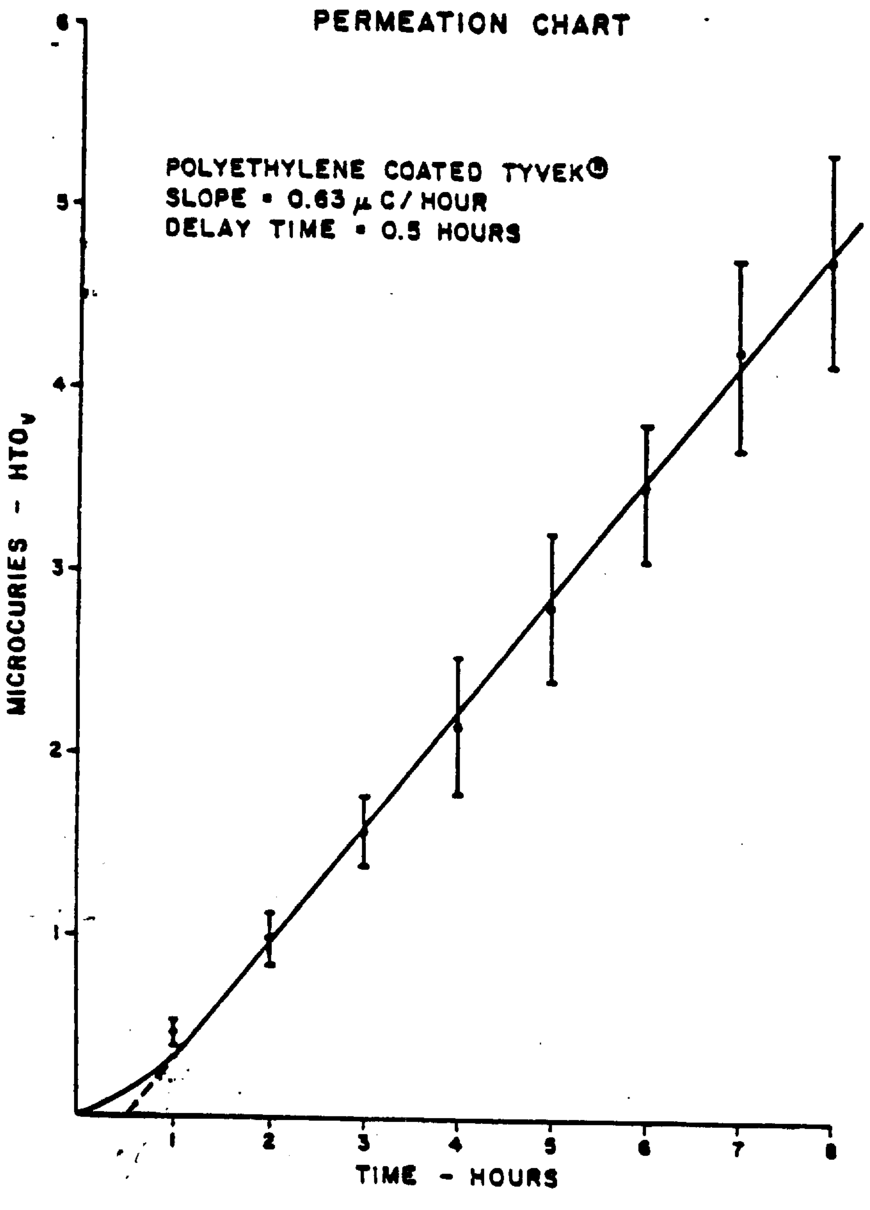

Figure 2 - Hageman Permeation Chart for Polyethylene Coated Tyvek

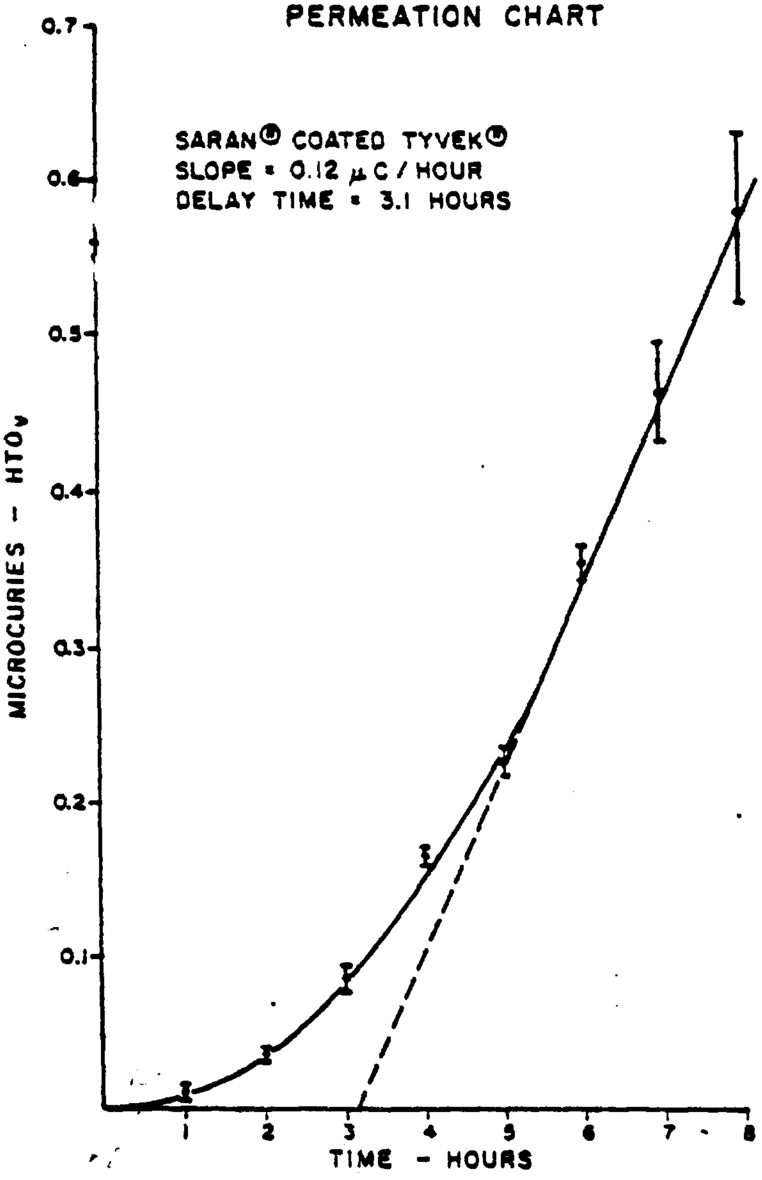

Figure 3 - Hageman Permeation Chart for Saran Coated Tyvek

TABLE VIII

PERMEABILITY CONSTANTS OF TRITIUM GAS THROUGH VARIOUS ELASTOMER AND POLYMER MATERIALS BY STEINMEYER AND BRAUN (1975)

Material	Thickness (cm)	Permeability Constant P $\frac{(cm^3)(cm)}{(cm^2)(sec)(atm)}$ at ~25°C
Latex	0.023	2.58×10^{-7}
Buna-N (Nitrate Butadiene rubber)	0.032	4.4×10^{-8}
Teflon (polytetrafluroethylene)	0.007	1.01×10^{-7}
Kapton	0.009	1.86×10^{-8}

TABLE IX

MATERIAL CHARACTERISTICS OF HT AND HTO IN SOME ORGANIC MATERIALS BY GAEVOI (1966)

Material	Permeability $\frac{(liter)(Ci)(cm)}{(cm^2)(sec)(Ci)}$ HT	HTO
Natural rubber	3.3×10^{-10}	2.3×10^{-9}
80/277 PVC formulation	1.2×10^{-10}	1.3×10^{-8}
L-7 Nairite latex	9.3×10^{-11}	3.7×10^{-10}
Polyethylene (à = 0.92 g/cm^3)	5.3×10^{-11}	4.5×10^{-10}
Teflon-2b	1.7×10^{-11}	1.0×10^{-9}
Terylene	5.6×10^{-12}	3.8×10^{-10}
Perfol PK-4	4.6×10^{-12}	4.7×10^{-10}

Connaley,[14] of Radian Corporation performed tritium permeation tests on several materials for Savannah River including an 8 mil Dow-Saran Research experimental film and several glove materials. (This report will be sent to Los Alamos when available.) The Dow-Saran film is a layered material of chlorinated polyethylene (CPE), Saran, and ethylene vinyl acetate. This material can be heat sealed which has been a problem with prior Saran component films. The materials were tested with tritiated solvents and HTO. Radian used a 10 ml permeation cell and liquid scintillation counting. An 8 cm^2 film sample was challenged with an HTO concentration of ~5-10 mCi/ml. The CPE-Saran material had a breakthrough of six times longer than the old PVC. The permeation rate for CPE-Saran is 1/4 that of PVC.

TABLE X

PERMEABILITY DATA OF GLOVE MATERIALS BY DOUGHTY AND WEST (1981)

Sample	Thickness (in.)	Temp. (°C)	Relative Humidity	HTO_V Permeability $\frac{(cm^3)(cm)}{cm^2\ sec\ TORR}$
Butyl Rubber	.020	39	78	8.6×10^{-10}
Butyl	.020	39.5	84	6.6×10^{-10}
Neoprene	.018	39	83	$161.\times10^{-10}$
Hypalon	.017	38	80	$113.\times10^{-10}$
EPDM/ Butyl	.035	37.2	69	7.3×10^{-10}
Hypalon/ Neoprene	.035	38	65	114×10^{-10}

(Breakthrough is that point at which the permeant first appears on the downstream side of the material. The permeation rate is the rate at which the permeant appears.) The CPE-Saran material was chosen for the construction of the new Savannah River tritium suit.

C. Compilation of Data from III-B

Table XI is a compilation of permeability constants from the reviewed documents. The data were reduced to common units and tabulated by material.

TABLE XI

POLYVINYLCHLORIDE (PVC)

Reference	Thickness (cm)	Permeability Constant $\frac{(cm^3)(cm)}{(cm^2)(sec)(cm\ Hg)} \times 10^{-8}$ HT	HTO
Hughes, 1962	0.036	0.148 (25°C)	
Billard, 1968	0.03	1.3	>70
	0.05	1.1	>40
Katoh, 1968			
Films	0.009		0.017
Decon Suits	0.019		0.026
Airline Suits	0.031		0.029
POLYETHYLENE (PE)			
Katoh, 1968	0.0065	0.000238	
SARAN			
Billard, 1968	0.009	0.03	0.7
	0.005	0.02	0.3
Charamathieu, 1970	0.009	0.03	0.16
TEFLON (POLYTETRAFLOUROETHYLENE)			
Steinmeyer, 1975	0.007		0.133

TABLE XI (continued)

MATERIAL COMBINATIONS AND LAMINATES

Material	Reference	Thickness (cm)	Permeability Constant $\frac{(cm^3)(cm)}{(cm^2)(sec)(cm\ Hg)} \times 10^{-8}$	
			HT	HTO
Cotton-Neoprene	Billard, 1968	0.042	4.5	>4.00
EPDM/Butyl	Doughty, 1981	0.089		0.73
Hypalon/Neoprene	Doughty, 1981	0.089		11.40
Nylon-Butyl	Billard, 1968	0.046	0.4	0.20
Nylon-PVC	Billard, 1968	0.037	0.23	>5.00
Saran- PE	Charamathieu, 1970	0.0089		0.42
PVC-Saran-PVC	Charamathieu, 1970	0.0419	0.11	1.81
Laminate		0.0211	0.09	1.20
Saran - PE	Billard, 1968	0.008	0.10	1.50
Saran - PE	Billard, 1968	0.011	0.06	1.20
Saran - PE	Charamathieu, 1970	0.0089	0.03	
Saran - PVC Laminate	Charamathieu, 1970	0.069		0.43

RUBBERS

Material	Reference	Thickness (cm)	HT	HTO
Buna-N (Nitrate butadiene)	Steinmeyer, 1975	0.032	0.0579	
Butyl	Doughty, 1981	0.051		6.6
(iso-Butylene)	Doughty, 1981	0.051		0.86
Technibutyl	Charamathieu, 1970	90.104	0.71	
		0.083		0.36

TABLE XI (continued)

NEOPRENE

	Thickness	Permeability Constant $\frac{(cm^3)(cm)}{(cm^2)(sec)(cm\ Hg)} \times 10^{-8}$	
Reference	(cm)	HT	HTO
Hughes, 1962	0.068	0.118 (25°C)	
Katoh, 1968	0.56		ä0.004
Billard, 1968	0.064	1.20	4.30
Charamathieu, 1970	0.069	0.70	
	0.074		1.10
Doughty, 1981	0.046		16.10

LATEX

Katoh, 1968	0.37m/m		~0.001
Billard, 1968	0.06	4.40	40.000
Steinmeyer, 1975	0.023	0.340	

OTHER MATERIALS

		Thickness	Permeability Constant $\frac{(cm^3)(cm)}{(cm^2)(sec)(cmHg)} \times 10^{-8}$	
Material	Reference	(cm)	HT	HTO
Butasol	Billard, 1968	0.039	0.40	0.80
Hypalon (Chlorosulfonated Polyethylene	Doughty, 1981	0.043		1.13
Kapton (polyimide)	Steinmeyer, 1975	0.009	0.025	
Aluminized Mylar	Billard, 1968	0.0025	0.002	0.50
Nylane	Billard, 1968	0.0085	0.30	1.90
Layered Polyester	Charamathieu, 1970	0.145	0.02	
Polyurethane	Charamathieu, 1970	0.0251	0.57	
Terphane	Billard, 1968	0.025	0.14	4.20
Terthane 082	Billard, 1968	0.008	0.16	>2.00
Terthane 6A2	Billard, 1968	0.013	0.14	1.70
Terthane 6T2	Billard, 1968	0.0075	0.10	1.10
Terthene	Charamathieu, 1970	0.008		1.10
Crystallized Vinyl	Billard, 1968	0.046	0.80	>30.0
Crystallized Vinyl	Charamathieu, 1970	0.046		15.0

2. Discussion of Table XI. There is generally good agreement among the investigators measuring PVC and Saran. Measurements on other polymers or laminates have not demonstrated such good agreement. The polymers with the lowest permeability are normally those exhibiting a high degree of ordered structure. Comparisons of the data presented in Table XI show that Saran is much less permeable than PVC. Laminates of PVC-Saran-PVC, Saran PE, and Saran-PVC are show less permeability than PVC. Latex and butyl rubber are less permeable than neoprene.

D. Garment Tests

In 1958, Butler and Van Wyck[15] of Savannah River Plant tested a one-piece 12 mil PVC suit and a two-piece 6 mil PVC suit with HTO_v to determine their integrity. They also laundered a one-piece suit and tested it to determine changes in protection, and learned that laundering does not appear to affect the overall integrity of the PVC. The one-piece suit in all tests provided better protection than the two-piece, as one would expect. When the air activities in the test box and in the suit were plotted against time of exposure for each specific test, all curves were similar in shape and showed two characteristics:

1. There was a time lag of 5 to 10 min before tritium within the box began to penetrate the suit. This lag did not appear to vary with the activity in the box or with the PVC thickness. The time lag offers a protective safety factor if the suits are exposed to sudden bursts of activity.
2. The maximum or saturation activity of the air within the suit was achieved in 20 to 30 min after peak activity was reached in the box.

To provide a guide for personnel exposure control, the data were transposed to a graph (Fig. 4) which showed the time in which 1 mCi of tritium would be assimilated when wearing either suit in a specific concentration. This graph served as a guide to determine which suit should be worn and the working time limit for that suit. Data from these tests have provided an effective method of controlling personnel exposures, and have permitted extensive use of the less expensive two-piece suit (cheaper by a factor of 7), resulting in a considerable cost reduction.

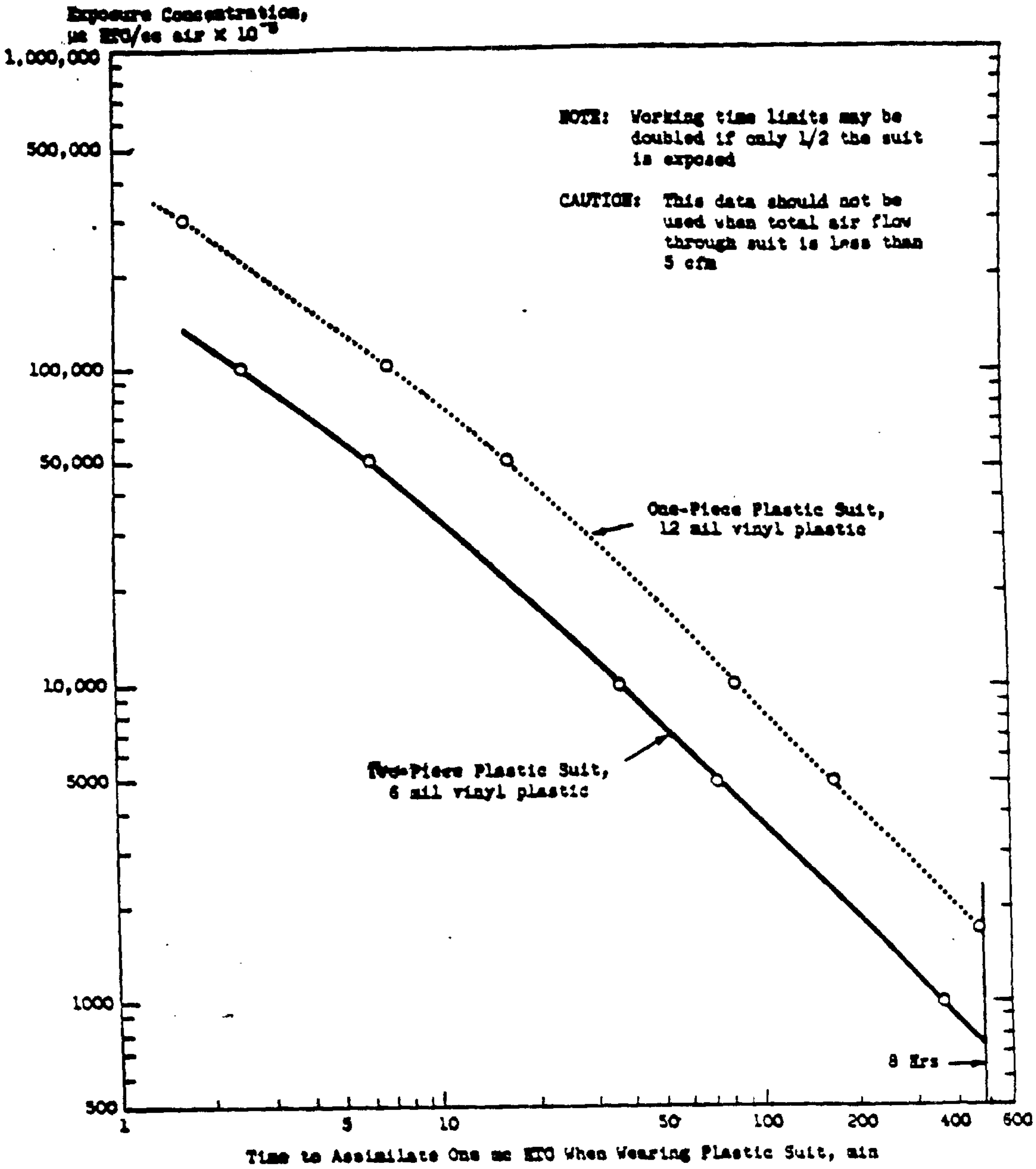

Figure 4 - Time Limits for Plastic Suit Use in Tritium Oxide Atmospheres at Savannah River Plant

Katoh (1968)[7] evaluated airline suits at two ventilation rates. Suit A designed by the Japanese Atomic Energy Research Institute (JAERI) gave a safety factor SF = HT Conc._{in}/HT $\text{Conc.}_{\text{out}}$ x 100 per cent of 0.34 per cent at the higher of flow (226 l/m) compared to 0.45 per cent at the lower flow of 170 l/m. Suit B designed by protective clothing committee in Japan, gave a SF of 0.12 per cent at the higher flow and 0.16 per cent at the lower flow. Katoh reported that the tritium exposure to a working man wearing a suit decreased by a factor of 200-1000 which he indicated agreed with Osborne and others.

Billard and Charamathieu (1968)[16] conducted a study to determine the efficiency of French protective clothing in a tritium atmosphere. They compared and rated nine suits. Only one of the nine suits was rated "Tres bonne" or very good. It was a one-piece suit with a tight helmet. The authors indicated that the suit's air distribution was bonne (good). The efficiency at the respiratory tract (at 30 l/min) for the new suit was >16,000 and for a used suit was 750. This suit was tested a second time, and the results confirmed the first. Later that year, Charamathieu[17] published "Le Tritium En Radio protection" in which he presents HTO measurements for various protective garments. Table XII presents these data.

Wittenberg (1977)[18] reported on the successful operation of a glovebox containment system when 1.75 X 10^4 Ci of tritium was accidentally released to the glovebox atmosphere. The total gaseous release to the environment was 2.2 Ci. The tritium concentration in the body fluids of the only worker increased by only 2 μCi/l. The appearance rate of tritium in the room and the absorption of tritium by the worker were adequately described by permeation calculations for the molecular species of T_2 and HTO through the butyl rubber gloves.

IV. SURVEY OF PROTECTIVE CLOTHING USE

A. Survey Methodology

The list of DOE tritium handling facilities to be surveyed was developed at Los Alamos, and confirmed with Ray Cooperstein, the DOE

TABLE XII

RESULTS OF GARMENT TESTS (Charamathieu-1968)

Protective Garments	HTO Conc. at Suit Exterior $\mu Ci/cm^3 10^6$	Duration Exposure (min)	HTO Conc. Within Suit $\mu Ci/cm^3 10^6$	Internal Contamination of person = A μCi (a)	Effective Period Days	Calculated Absorption w/o protection B	Efficiency B/A	Suppl Ai (b
Cotton Overalls w/o Ventilation	50	120		200		240	1,2	
	600	10		40-80		240	3-6	
Mask only, w/o Skin Protection	800	20		85	8	640	7,5	
One-Piece Garment In Butyl Rubber	5000	45		25		9000	360	50-
	5000	27-45		10		7800	>800	50-
Two-Piece PVC Garment with Supplied Air Mask	600	20		<25		480	>20	
	5000	15		35	6-13	3000	90	
		15		40	7-13	3000	80	
		18		45	8	3600	80	
		15		40	9	3000	80	
		10		25	6-10	2000	80	
		10		15	11	2000	130	
		9-15		<10		2000	>200	
Two-Piece Garment In Butyl Rubber With Supplied Air Mask	600	20		<15		480	>30	
	800	20		<15		640	>40	
One-Piece PVC Garment	600	90	<7	<15		2200	>140	70-1
	2400	90	<90	85	5	8600	100	85
	2800	80	<110	65		9000	140	60
	3000	120	<140	<20		14000	>700	65
	7500	70	<140	65		21000	320	80
	18000	45	35	40	4-13	32000	800	45

(a) < = Below detection
(b) Due to expermental error, the listed values are too large, and indicated by the sine "<".

(1366Z)

administrator for these facilities. The survey questionnaire was developed following five site visits to DOE tritium handling facilities and determining the information required for the survey. Results were compiled by phone input. Each facility representative was asked for the following: (1) the tritium hazard or hazards, (2) the potential tritium concentration or exposure, (3) the frequency of tritium bioassay (urinalysis), (4) the personnel protective equipment in use, and (5) test results for their equipment if available. The information in this survey is presented as it was obtained from the contacts. Ten DOE contractor facilities (Appendix A) and two Canadian facilities (Appendix B) were surveyed.

B. U.S. Department of Energy Facilities

DOE regulations for tritium exposure are (1) the maximum permissible concentration of tritiated water in air (MPC_a) for a 40-hour work week is 5 $\mu Ci/m^3$ and (2) for molecular tritium the MPC_a is 20,000 $\mu Ci/m^3$. The maximum permissible body burden (MPB_B) is ~1000μCi, or with a water content of 43 kg, about 0.023 $\mu Ci/cm^3$ (23 μCi/l). This quantity of tritium delivers a dose-equivalent rate of 0.1 rem/week, or if maintained for 1 year, 5 rems/year. A single exposure that brings the body water to a concentration of 1 $\mu Ci/cm^3$ causes a total dose equivalent during the time of elimination of about 9 rems.[1]

1. Los Alamos National Laboratory.

The only significant personnel exposure at Los Alamos tritium systems result from inhalation and skin absorption of HTO. In all systems there is a small amount of HTO present on metal surfaces, in oils, on glassware, or as a vapor with the HT.

a. Los Alamos Tritium Sampling Criteria.

Workers covered by this program are those who either regularly or intermittently work on systems that contain 1 Ci or more of tritium or who work in decontaminating those systems. The basic routine sampling program requires that these workers submit spot urine samples for analysis on a schedule of once every 2 weeks. More frequent urine samples are required following significant exposures.

Criteria for Suspected Exposures. Tritium workers who recognize that they may have been exposed because of equipment failure, breach of procedures, instrumentation alarms, or high swipe counts are candidates for special sampling. Procedures for sample submission are dependent upon the suspected seriousness of the exposure. An exposure can occur when:

- tritium-contaminated vacuum pump oil has been spilled on bare skin or clothing,
- highly contaminated material or equipment is handled,
- a tritium instrument alarms, or
- working in an area without rubber or plastic gloves and tritium swipes are above 10 μCi.

Los Alamos procedures for monitoring tritium exposures require the worker to give an initial urine sample immediately after he/she exits the area and spot samples 2 hours after initial. The follow-up sampling schedule depends on the initial and spot sample results.

Previous Sample Result (μCi/l)	Bioassay Procedures
>100	Submit daily samples including weekends and holidays, preferably at 24-hour intervals.
10 to 100	Submit weekly samples.
1 to 10	Submit next sample within 1 month of the time of the previous sample, UNLESS on a routine sampling program (every 2 weeks) or UNTIL the next suspected exposure.
< 1	Submit no more samples until the next suspected exposure, or until the next routine (every 2 weeks) scheduled sample.

b. Tritium Handling Facility (THF). THF was activated December 1974 and designed to handle large quantities of tritium in the form of metal tritides or gas. The system consists of an 11.5 m^3 dry-box line, associated gas purification system, and an effluent treatment system.[19]

The potential HT concentration at the THF is ~6 Ci/m^3. Workers submit urine samples for tritium bioassay once every 2 weeks unless an exposure is believed to have occurred. The average THF worker exposure based on tritium urine assay was 0.45 rem in 1985. Norton 30 mil Hypalon gloves (chlorosulfonated polyethylene) are attached to the THF dry boxes. The worker wears a pair of Handguards, Inc. 1.75 mil shoulder-length gloves, covered by Pioneer Quixam Pylox V-5 PVC gloves. In some cases, cotton liners are worn against the hands. A complete glove change every 20 minutes of work helps control worker exposure at THF. When routine maintenance is performed and HTO is not anticipated, the worker wears coveralls, Pylox PVC gloves, and the Mine Safety Appliance (MSA) Self Contained Breathing Apparatus (SCBA). The two-piece Los Alamos suit, described later in this section, is available for emergency.

c. Tritium Systems Test Assembly (TSTA). TSTA, dedicated October 1982, was designed for a large quantity (150 gm or ~15 x 10^5 Ci) of HT in the inventory. Protection is based on a methodology of containment, detection, and recovery.[20] At the time of the survey, TSTA had 30 gm (~3 x 10^{5}Ci) HT in the loop. Potential exposures range from 20-100 mCi/m^3 depending upon the experiment being conducted at the time. Based on tritium urine bioassays, the greatest exposure in 1985 was 0.02 rems for TSTA. Norton 30 mil Hypalon gloves are attached to the dry boxes and the worker wears Pioneer Quixam Pylox V-5 gloves over a pair of Handguards Inc. 1.75 mil shoulder length gloves. Cotton liners are optional. The Los Alamos suit is also available.

d. WX-5 Facility. The WX-5 facility handles varying quantities of HT which correspond to the operation at that time. Sometimes as much as 12 to 18 gm HT or 12 X 10^4 - 18 X 10^4 Ci are required. If any equipment malfunctions or a line ruptures, a portion of the HT can be released into the room where the operation is being performed. The room concentration normally measures 0.5 μCi/m^3 during operations. In 1985, the two principal workers received 1.40 and 1.07 rem, determined by urine assay. Daily work is performed with the worker wearing Pylox V-5 PVC gloves that are normally changed every 5 minutes. The Los Alamos suit is available whenever it is required. A new facility for this operation is being designed/constructed with sophisticated ventilation systems and controls.

e. Los Alamos Suit. When an HTO emergency occurs or is anticipated, the Los Alamos two-piece, airline supplied-air suit is worn. This device was tested by Los Alamos in 1979 and accepted by DOE.* It is constructed of 6 mil PVC and consists of a slipover jacket with sealed-on hood. The trousers have sealed-on booties. The gloves of this suit are taped to hard cuffs that are sealed to the sleeves of the jacket. An aerosol protection factor (PF_a) $\geq$10,000 was determined for the suit at a 6 cfm airflow. Los Alamos has not conducted tritium permeation tests on this suit material.

2. Monsanto Research Corporation - Mound Laboratory. The common tritium hazard at Mound is HT, with few instances of HTO. Jay Doty, Manager of Health Physics, indicated that lead-lined/coated Norton Hypalon gloves were used on the Mound dry boxes. The worker wears cotton liners and latex surgeons gloves or orange rubber gloves. Each worker is requested to submit a urine sample twice each week for bioassay. The laboratory alerts Health Physics when a worker's sample is $\geq$10 µCi/l. The average worker's bioassay is <1 µCi/l. Ray Brashear, Health Physics, said that in their facility they strive to reduce the HT concentration < 1800 $\mu Ci/m^3$ before the area is entered wearing the present bubble suit.

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*The Personnel Protection Studies Section of Los Alamos is under DOE contract to conduct rigorous testing of nonapprovable equipment such as full suits, to determine if the equipment will adequately protect the user. All testing is performed according to the "Acceptance-Testing Procedures for Air-Line Supplied Air Suits" [LA-10156-MS (1984)] and the licensee's SOP submitted with the equipment. The test results are then considered by the Respirator Advisory Committee members, who make recommendations for acceptance to DOE. DOE has the final acceptance responsibility.

In 1973,[21] a three-man committee at Mound Laboratory evaluated glove sets to be used on bubble suits for tritium operations using criteria of resistance to tritium permeation, toughness, color, and feel. A four-glove concept was decided upon in order to achieve maximum personnel hand protection. The employees regularly wear a cloth liner and 7 mil natural-rubber glove. The committee recommended the mandatory use of Buna-N gloves due to its extreme strength and resistance to tritium permeation. The first glove set recommended was the combination of butyl, Buna-N, and natural rubber because this set provided a barrier with extreme resistance to tritium permeation, extreme toughness, contrasting colors, and an adequate sense of feel. The only restriction for this set was the low resistance of the butyl glove to organic solvents. The second recommended set consisted of two natural-rubber gloves and one Buna-N.

The 6 mil PVC single-use, disposable bubble suit presently being used for tritium and other radiological hazards at Mound was tested at Los Alamos in 1973 and accepted by DOE. A new single use, disposable suit being considered, is also constructed of 6 mil PVC but has an improved design which has borrowed the good points of both the Idaho Falls and Savannah River suits. According to Mound personnel the old suit provides a PF_a ~1800-2000. Independent (non-DOE) tests on the new suit report a $PF_a \geq 10,000$ at an unknown airflow. The new suit will accommodate a hard hat and the top will have an apron for easier bending by the wearer. It has reinforced knees and booties in the pants and vented arms and legs. The Idaho Falls air-distribution system is used in the suit and provides a reduction in noise level. Doty indicated that due to the high estimated cost of ~$300/suit, the decision to continue the development and manufacture of the new suit will depend upon Mound's need for improved protection and available funds.

<u>3. I. E. Du Pont de Nemours and Company - Savannah River Plant (SRP)</u>. The potential HT hazard at SRP is as high as 10,000 Ci/ml (1×10^{-7} Ci/l). Tritium workers submit samples for bioassay on a varied schedule, which depends upon the job.

If personnel are required to enter a process hood (dry box) to perform maintenance, each person participating in the work submits one urine sample each work day for bioassay. If there has been unexpected activity detected during a maintenance job, then all personnel involved submit samples 90 minutes after the potential exposure. Personnel regularly assigned to tritium areas submit urine samples once each week. Office personnel working in the building who are not regularly in the tritium areas, submit samples once each calendar month. Bioassays are also performed whenever there is a request from any of the health physics staff.

SRP health physics personnel monitor for tritium activity at several points during a job by placing sampling lines: (1) on the exterior of the suit, front adjacent to the face and breathing zone of the wearer; (2) at the point of break in the hood system to determine the activity being released; (3) inside the hood being repaired, and (4) in the room where the hoods are located.

North (new name for Norton) neoprene gloves are used on the SRP dry boxes. The worker wears cotton liners and latex gloves with coveralls. SRP has recently had Radian Corp., Austin, Texas, perform tritium permeation tests on butyl rubber and other glove materials.

The present single-use, disposable suit being worn at SRP is constructed of 12 mil PVC and is of the same design as the 6 mil suit that was tested at Los Alamos in 1980 and accepted by DOE. The supplied-air suit is a two-piece design consisting of a long sleeve pullover jacket with a clear plastic helmet and separate pants with boots sealed to the legs. The protection factor determined for the 6 mil suit was $PF_a \geq 10{,}000$ at an airflow of 18-20 cfm. SRP uses approximately 10,000 suits/year in their operations.

The new SRP suit, recently submitted to DOE for testing, is constructed of an 8 mil experimental Dow-Saran Research film that can be heat sealed. The film is a laminated material of chlorinated polyethylene (CPE), Saran, and ethylene vinyl acetate (EVA). The design of the new suit is the same as the 1980 suit.

Radian Corporation performed the tritium permeation tests on several materials for SRP as well as ~ ten glove materials. The materials were tested with tritiated solvents and HTO_v. (This report is to be sent to Los Alamos when available and will be forwarded to DOE.)

4. General Electric Company's Pinellas Plant (GEPP). Rich Greene, GEPP health physicist, indicated that the tritium exposures were fairly low, with 500 mrem/year for the maximum exposure. Norton Butasol-butyl 15 ml gloves are used on the GEPP dry boxes with the worker wearing 5 mil Pylox V-5 PVC gloves. Each tritium area worker submits a sample for bioassay once a week. The laboratory reports any amount >0.1 μCi/l (0.85 mrem) to the health physics department. Vacuum-system maintenance personnel who are at the highest risk use 5 mil Pylox V-5 PVC gloves for low contamination items such as tools, etc., but wear latex gloves over the PVC gloves with 8 mil PVC sleeves for higher contamination levels and HTO_v. Full-facepiece supplied-air respirators (Scott or MSA) are used if any airborne tritium is detected. Nonroutine workers submit a urine sample for tritium bioassay on the day of exposure.

5. Lawrence Livermore National Laboratory (LLNL). Harry Howe, Jr., health physicist for the LLNL tritium facility, said that most of the LLNL tritium work is with HT in dry boxes with attached Norton Hypalon gloves. The worker wears cotton liners, Pioneer Quixam Pylox V-5 (5 mil) PVC gloves, and plastic arm-length gloves. The three glove layers are changed every 15 minutes. The potential tritium hazard is low. Howe reported the LLNL 1984 total personnel tritium exposure was 479 mrem for 52 workers. Twenty-eight (28) people had no detectable dose with 24 receiving a detectable dose. The highest 1984 individual exposure was 122 mrem. In 1985 the total personnel exposure to tritium was 507 mrem for 51 workers, 24 receiving no detectable dose and 28 having detectable doses. The highest individual exposure was 75 mrem in 1985. LLNL tritium workers submit urine samples for tritium bioassay once a week. LLNL does not have a plastic suit or respirators available for emergencies; however, this equipment is being considered.

6. Sandia National Laboratories, Livermore, California. Don Wright, Sandia health physicist, said that workers wore cotton glove liners and Pioneer Quixam Pylox V-5 PVC gloves inside the Hypalon dry box gloves. The highest potential HT hazard at Sandia is 120gm or ~12x 10^5 Ci. The workers submit samples for tritium bioassay on a weekly schedule, unless an exposure occurs, then more frequently as dictated by the situation. The suit available for emergencies is the old Mound PVC suit. Don is investigating buying new suits and is considering the new CPE-Saran, SRP suit.

7. Rockwell International's Rocky Flats Plant, Golden, CO (RFP). Richard Link, RFP Health Physics Group Leader, said there was very little tritium work being done at RFP at present, and their tritium hazard is in the few Ci range. HT is handled in dry boxes with recirculating gas control. They use 45 mil Hypalon dry box gloves constructed of two layers of Hypalon with sandwiched layer of neoprene. Workers only wear cotton glove liners in one operation to cushion the material being handled. One case of tritium permeation in the pCi range has been seen at RFP. The tritium workers have monthly bioassay or in case of an exposure, one urine sample is collected 24 hours after the incident.

Suits are not used because RFP health physicists have calculated that the worst possible case of tritium contamination would be in the ~10 Ci range. The only documented amounts of tritium contamination have been amounts less than the DOE regulation of 5 $\mu Ci/m^3$.

8. Oak Ridge National Laboratory (ORNL). Hal Butler, ORNL health physicist indicated that most of their work is conducted in hooded operations with excellent airflows. ORNL purchases tritium from SRP and retanks it for sale to commercial concerns. They also fabricate tritium accelerator targets ranging up to 100 Ci. Twelve mil (0.012 in.) Safeco Corporation natural-latex surgeons gloves, cotton liners, and Remco shoulder length 0.020 in. butyl-rubber gloves are used at ORNL. Norton Hypalon 0.015 in. gloves are used on the dry boxes. ORNL tritium workers provide samples for weekly bioassays and following each suspected incident.

In operations such as target fabrication, when ORNL workers must use a full suit, they use the ORNL suit tested at Los Alamos in 1982 and accepted by DOE. This two-piece suit is constructed of 6 mil PVC. The measured protection factor was a $PF_a \geq 10,000$ at 8-9 cfm airflow.

9. Westinghouse Idaho Nuclear.

10. Argonne National Laboratory, Idaho. Idaho Nuclear and Argonne West were both contacted but representatives reported that each facility had negligible amounts of tritium and no formal programs for tritium monitoring or protection.

C. Canadian Facilities

Two Canadian Facilities (Appendix II) were contacted regarding equipment used for protection against tritium. The Canadian maximum permissible concentration is $MPC_a = 10\ \mu Ci/m^3$.[22]

1. Ontario Hydro, Pickering, Ontario, Canada. The tritium hazard at Ontario Hydro is HTO_v and HTO_l. Typical concentrations for the Pickering Generating Station are 2-10 MPC_a with short-term concentrations as high as 900 MPC_a in accessible areas. Average concentrations in access-controlled areas (boiler room and moderator room) are normally < 100 MPC_a with short-term concentrations of several hundred MPC_a. Personnel wear full protective clothing in the access-controlled areas. The average tritium dose per worker is 0.2 man-rem/MPC_a.[22]

Regular bioassay samples are required at stations where tritium exposures are more or less chronic. Sampling frequencies adopted in Ontario Hydro are as follows:

Frequency	Work Group
Weekly	Personnel who regularly perform radioactive work involving radiation exposure (e.g. operators, maintenance staff, chemical technicians).

Monthly	Personnel based in the Radiation Area who normally do not perform work involving radiation exposure (e.g. health physicists, engineers, supply staff).
Quarterly	Other station staff.

In addition, nonroutine bioassay samples are required following known or suspected exposures. These samples are submitted about 2 hours after the exposure to permit the tritium to equilibrate in body fluid.

Bioassay control levels have been established in Ontario Hydro to limit exposures and help achieve as low as reasonably achievable (ALARA) objectives. These levels differ slightly from station to station. Levels adopted at Pickering are as follows:

Restriction	25 μCi/l
Removal	50 μCi/l

Restriction and Removal means that no radioactive work may be performed unless a Radiological Work Plan has been approved. In the case of the Restriction category, the plan must be approved by the individual's supervisor, and in the case of Removal by the duty Shift Supervisor. All staff are encouraged to stay below a burden of 10 μCi/l.

John Stephenson of the Ontario Hydro Safety Device and Protective Clothing Section, said that the primary personnel protection for tritiated heavy water are full suits developed at Ontario Hydro. The reusable "neck entry" Mark III is constructed of a laminate of a 2.5 oz/yd^2 nylon fabric base with 40 threads/in^2 in both directions. The nylon is then covered with 4 mil PVC on the inside and 6 mil PVC on the outside, making the total material weight of 13 oz/yd^2. The Mark IV has a similar nylon fabric base with neoprene rubber coatings on both sides, the final fabric weight is ~13 oz/yd^2. Permeation testing was conducted by the Ontario Hydro Research Department following the ASTM E96-80[23] standard method for Water Vapor Transmission of Materials.

The water-permeation measurements derived from testing the materials are as follows:

PVC-nylon	$P = 27\text{gm/m}^2\text{/24 hrs.}$
Neoprene-nylon	$P = 6\text{gm/m}^2\text{/24 hrs.}$

The Mark III and Mark IV protection factors (PF_t) were measured in a tritium atmosphere* in 1978. The results were $PF_t \geq 1000$. Stephenson said that PF_ts were measured again in 1980 when the Mark III design was improved. Further measurements were made in 1984 on used Mark IIIs and the resulting $PF_t \geq 500$. Measurements were then made on new Mark IIIs and again the $PF_t \geq 500$. Stephenson has not determined the reason for these PF_t differences, but indicated that tritium permeation tests rerun in 1984 gave results very close to those measured in 1978. Stephenson also said aerosol protection factors conducted on both the Mark III and IV give $PF_a \geq 2000$ to 2500.

a. Tritium Environmental Chamber. Ontario Hydro has the only tritium environmental chamber in which suit protection factors are measured while a human test subject wears the suit in a HTO_v atmosphere. The chamber dimensions are 12' X 10' X 8' with an attached 4' x 4' airlock. The airflow to the chamber is 50 cfm. When testing suits, 150 MPC_a HTO is used. Ontario Hydro personnel look for 0.1-0.3 µCi/l tritium in urine above the baseline. (Ontario Hydro suit testing procedures for the tritium environmental chamber tests are presented in Appendix 2.)

2. Chalk River Nuclear Laboratories (CRNL).

The tritium hazard at CRNL is also HTO_v and HTO_l. The major sources of tritium exposure are found in the NRU reactor and the heavy water upgrading plant.[22] Tritium concentrations in reactor-heavy water are up to 18 Ci/kg. Tritium concentrations in the accessible areas of the NRU research reactor range typically between 0.1-0.5 MPC_a. At the CRNL heavy-water upgrading plant, they average 0.1-0.2 MPC_a. These levels are

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* NOTE: In US PF testing, the amount of aerosol (not tritium) that enters the device is measured and the results are given in aerosol protection factors (PF_a). PF_a and PF_t can not be directly compared.

significantly lower than that experienced at Ontario Hydro's power reactors where high-temperature, high-pressure heavy water systems are present.

Stan Linauskas said that the reusable CRNL air-cooled suit is used for all hazards, tritium and radioactive particulates. It is a three-piece garment: a 12 mil clear PVC long jacket and high waders with a bib front with several different shaped hoods. The smaller, soft hoods can be worn in confined spaces where the common hood is 12 mil PVC hard plastic, somewhat like the old motorcycle helmet. The air-distribution system has hoses down the legs and arms with a distribution plenum on the belt at the waist. Airflow is 25 cfm at the distributor. Linauskas said that for shorter use time, a hood with airflow to the head is used.

CRNL also has a disposable 6 mil one-piece PVC suit with a neck opening. This suit can be worn with an airline respirator with a hood or an air-supply yolk around the neck. The air-supply can be either a downflow or an upflow design.

Richard Osborne, Head of the CRNL Environmental Research Branch, conducted tritium permeation tests with the 6 mil suit several years ago. The results of these tests were not available.

CRNL does not assign specific protection factors to the suit because of its diverse use, however, if the job is to be performed in a low tritium concentration area; i.e., 500-700 MPC_a, then CRNL personnel would use the 6 mil disposable suit.

3. Canadian Fusion Fuels Technology Project (CFFTP). K. Y. Wong, CFFTP said that the CFFTP had been developed to serve any country with operating fusion reactors; i.e., European, Japanese, and United States. He indicated that Canada will not have a fusion reactor on line for a number of years.

When questioned about the protective equipment that a Canadian tritium worker wears, Wong said that it depended upon the hazard and the time the worker would be in the hazard area. For example, with a high gamma of 5rem/hour, the worker would wear a respirator and not a bulky plastic suit. In the Canadian deuterium uranium (CANDU) stations, supplied-air plastic suits must be worn in certain designated

TABLE XIII

Facility	Suit Type	Material	Tritium Challenge	Protection Factor a= aerosol, t= tritium
Department of Energy				
Los Alamos National Laboratory	2 piece Air-supplied (A-S) Respirator/ coveralls	6 mil PVC	HTO HT	$PF_a \geq 10,000$ (DOE)
Mound Laboratory	Present suit 2 piece A-S	6 mil PVC	HT/HTO	$PF_a = \sim 1800-2000$
	New suit 2 piece A-S	6 mil PVC	HT/HTO	$PF_a \geq 10,000$
Savannah River Plant	Present suit 2 piece A-S	12 mil PVC	HT/HTO	$PF_a \geq 10,000$ for 6 mil suit (DOE)
	New suit 2 piece A-S	8 mil CPE-Saran	HT/HTO	Submitted to DOE/86
Pinellas Plant	none (Dry Box)	Air-supplied respirator	Airborne HT	
Lawrence Livermore National Laboratories	none (Dry Box)		HT	
Sandia National Laboratories-Livermore	2 piece A-S Mound	6 mil PVC	HT	$PF_a = \sim 1800-2000$
Rocky Flats Plant	none (Dry Box)		HT	
Oak Ridge National Laboratory	2 piece A-S	6 mil PVC	HT	$PF_a = >10,000$ (DOE)
Canadian				
Ontario Hydro	Mark III, 1 piece neck entry A-S	PVC-Nylon-PVC	HTO	$PF_t \geq 500$ (OH) $PF_a \geq 2000-2500$
	Mark IV, 1 piece A-S	Neoprene-Nylon-Neoprene	HTO	$PF_t \geq 500$ (OH) $PF_a \geq 2000-2500$
Chalk River Nuclear Laboratories	3 piece Air-cooled suit	12 mil PVC	HTO	$PF_{a-hood} \geq 10.000$
	1 piece Air-cooled suit	6 mil PVC	HTO	

high-tritium hazard area such as moderator rooms or for certain jobs such as resin slurrying. In all other areas, plastic suits are mandatory if the tritium concentration exceeds 10 MPC_a or if the estimated exposure exceeds 10 MPC_a/hour. Their suit use philosophy is designed around user acceptance. The suit must be easily/quickly donned and removed. "A suit that requires 20 minutes to remove is not acceptable to a worker who is very tired, hot, sweaty, and ready to tear off any thing next to his skin," said Wong. The Mark III has air-cooling channels built in the suit and can be put on and zipped up. It can be donned in 2 minutes and removed in 1.5 minutes. The cost of the suit is $400 Canadian (~$300 US). The Mark III has a $PF_t \geq 1000$ (measured in a HTO_v environment in 1975) or possibly less in actual work. The suit air supply is 28 cfm. The Mark III can be laundred 100 times. The Mark IV can be dry cleaned. These suits are commercially available through Safco.

Wong said that organic tritium can be formed by metal catalysis of tritium and can be readily absorbed by the skin. CFFTP is considering supporting research to investigate this problem. Wong indicated that John Stephenson would play a major part in this research if it were funded.

D. Comparison of Suit Data

Table XIII was developed to collectively compare the suits, the measured suit PFs, and the tritium challenges at the facilities surveyed. Most of the facilities are using PVC suits. PF_a and PF_t cannot be directly compared.

PVC (6 and 12 mil) suits are being used by the DOE tritium handling facilities. Savannah River Plant has a CPE-Saran-EVA suit at Los Alamos for DOE acceptance testing. Ontario Hydro is using a PVC-Nylon-PVC suit (Mark III) and a Neoprene-Nylon-Neoprene suit (Mark IV) in their facilities.

V. TRITIUM PERMEATION TEST APPARATUS

A. Background

To determine the hazards caused by a toxic chemical permeating chemical protective clothing (CPC), two factors must be considered:

(1) breakthrough time (T_B), the time at which the permeant first appears on the downstream side of the material, and (2) permeation rate, the rate at which it appears. These two factors cannot be divorced completely from one another in determining the total threat posed. However, if no exposure is permitted, T_B is the important factor. If some exposure can be tolerated, the permeation rate is needed so that the total amount permeating after breakthrough can be calculated. In both cases, laboratory experiments must accurately simulate work conditions if safe working times are to be predicted.

The majority of the pertinent data cited in the literature review (Sec. III B) were permeability constants. These can be used to calculate the permeation rate by:

$$F = \frac{P(p1 - p2)}{L}$$

where F = Permeation rate
P = Permeability constant
p_1, p_2 = Pressure of permeating gas on upstream and downstream sides of the membrane
L = Membrane thickness.

These calculated permeation rates refer to the steady state and not to time immediately following breakthrough. Further, there are few data given on breakthrough times in the literature. Because of the lack of these data, it might be useful to repeat some of the earlier permeation testing to obtain T_Bs and permeation rates.

A more critical lack of data is that related to the permeation of substances other than HT, HTO_l, and HTO_v, such as tritiated organic solvents, oils and greases. In many operations, these compounds are necessarily involved if only in a subsidiary role, e.g., with vacuum and other fluid handling pumps. Many of these substances are nonvolatile so that once in contact with a clothing material they will preferentially permeate into, rather than vaporize from the material. These same substances may also be readily absorbed into the body.

Permeation rates for volatile permeants could be acquired using a permeation cell with a tritium detector built into the downstream side or by circulating the gas in the downstream side through an external detector. With water soluble compounds, water placed in the downstream side of the cell could be sampled periodically and analyzed for tritium. The presence of nonvolatile, nonwater soluble materials should be determined without having to remove them from the membrane surface. This could be accomplished most readily by employing a surface tritium detector. Although these counters usually have low efficiencies, they would probably be adequate because the tritiated permeant will be concentrated on the membrane. If not, a more sensitive detector could be designed and built.

B. Detectors

Discussions with Los Alamos personnel and others concerning tritium detectors indicate: there are detectors for determining quantities of HT and HTO, but there are very few that can be used to detect tritiated-hydrocarbons in small cells. A Japanese visitor invited to Los Alamos group in 1986-87, has worked in this area. His experience would be valuable in developing this type of detector.

VI. TRITIUM PERMEATION TESTING LABORATORIES

A. Commercial Laboratories

Southwest Research Institute, San Antonio, Texas and Radian Corporation, Austin, Texas, have or are performing small swatch testing for HT and HTO permeation as shown in the literature review. DOE should develop a program to evaluate these laboratories and any other commercial laboratories; as well as their equipment, testing methodologies, techniques, and resulting permeation data. This information would be valuable to DOE contractors and facilities requiring tritium permeation data.

B. DOE Test Laboratories

Two Los Alamos Laboratory sites have been identified that have appropriate controls for conducting tritium permeation tests and developing/testing surface detectors for the permeation of tritiated organic solvents, oils and greases.

Dan Doughty of Sandia National Laboratory, Livermore, California, indicated that his HTO permeation test system could be automated easily to eliminate the labor intensive aspect of the past testing.(13) Sandia would be very interested in testing materials proposed for tritium suits.

C. Other Tritium Research

Clay Easterly, Oak Ridge National Laboratory, indicated that he had recently completed work with EG and G-Idaho investigating the conversion of HT to HTO. He is presently writing up this data. He suggested that DOE should be interested in 'preventing the conversion of HT to HTO.' If this conversion could be controlled, worker protection would be simpler because HT is not as hazardous. ORNL would be interested in conducting further research in this area for DOE.

VII. CONCLUSIONS

A. Tritium Suits in use at DOE Facilities

The DOE tritium handling facilities are presently using PVC suits. There are other materials that exhibit lower tritium permeability constants than PVC, as can be seen in the literature review. However, many of these materials are not as mechanically adaptable to the end use of suits as PVC. PVC has low flammability, electrical resistance, and chemical resistance. It can be made into relatively light-weight suits with well sealed seams that are tear resistant and comfortable under normal use conditions.

Savannah River has had tritium permeation tests conducted on new materials and have chosen a laminate of CPE-Saran for their new tritium suit. This material is reported to have a breakthrough time of 6 times longer than the old PVC. The permeation rate for the material is 4 times less than PVC.

Other DOE facilities such as Sandia National Laboratory at Livermore, are interested in purchasing the new CPE-Saran, Savannah River suit for use in their tritium facilities.

VIII. RECOMMENDATIONS

A. We suggest that a topical meeting be held to allow representatives of each of the tritium handling facilities to discuss problems and solutions. During the survey, it was obvious that the facilities use printed literature developed by other facilities, but do not have the direct contact that could aid both parties with problem solving. DOE would benefit by determining the specific areas requiring its assistance.

B. The Canadian tritium environmental chamber should be visited and discussions held with the Canadians as to the possibility of a cooperative test agreement. In such an agreement, US DOE suits could be tested in the Canadian tritium chamber and a DOE facility, such as Los Alamos, could conduct aerosol protection factor testing for the Canadians. This would provide comparative data for the protective suits being used or developed in the US and Canada. This agreement would be mutually beneficial to both the US DOE and its facilities; as well as the Canadians.

C. Any commercial laboratories conducting tritium permeation testing for DOE facilities should be evaluated. This would provide DOE facilities with information regarding which laboratory could better provide the permeation data required.

D. Further research should be conducted to develop sensitive surface detectors for tritiated hydrocarbons such as vacuum pump oils.

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APPENDIX

APPENDIX A

The following DOE contractor facilities were visited (SV) or contacted by telephone (TC) regarding the protective equipment being used for personnel against tritium.

Los Alamos National Laboratory Los Alamos, NM (SV)	
Tritium Handling Facility	Joe Nasise
Tritium Systems Test Assembly (TSTA)	Jim Anderson, Roland Jalbert
TA-33 WX-5	Herman R. Maltrud
HSE-1 Tritium Monitors	Jose Gutierrez
Monsanto Research Corp.'s Mound Laboratory Miamisburg, OH (TC)	R. J. Rhude, Jay Doty, Ray Brashear, Herbert Stuart
E. I. du Pont de Nemours and Company's Savannah River Plant Aiken, SC (TC)	Christopher Wiernicke
General Electric Pinellas Plant St Petersburg, FL (SV)	Rick Greene
Lawrence Livermore National Laboratory Livermore, CA (SV)	Harry Howe, Jr., Nick Nicolosi
Sandia National Laboratory, Livermore, CA (SV)	Don Wright, Dan Doughty, and Ron Hafner
Rockwell International -Rocky Flats Plant Golden, CO (TC)	Richard Link
Oakridge National Laboratory Oakridge, TN (TC)	Clay Easterly, H. M. Butler, Mike Brooks
Westinghouse Idaho Nuclear Company Idaho Falls, ID	Jerry Bowman
Argonne National Laboratory (Idaho Site) Idaho Falls, ID	George Carr

Appendix B

The following Canadian facilities were contacted:

Ontario Hydro; Pickering, Ontario	John Stephenson, Michael Eisner
Chalk River Nuclear Laboratories Chalk River, Ontario	Stan Lanauskas
Canadian Fusion Fuels Technology Project	K. Y. Wong

Appendix C

1. Ontario Hydro Tritium Suit Testing Procedure

a. The standard suit testing procedure is outlined below:

(1) The test subject submits 10 urine samples for tritium assay over a 3-day period prior to the test to establish his baseline tritium level.

(2) Tritium concentration in the environmental chamber is established at the desired level by bubbling air through a heated (40°C) vessel containing tritiated water and uniformly distributing tritiated water vapour throughout the chamber using a circulating fan.

(3) The test subject enters the chamber in a protective suit with air supply to the suit maintained at the required level.

(4) The subject carries out an exercise and work protocol lasting approximately 2 h with rest periods at regular intervals. The exercises consist of:

(a) Walking on a treadmill (5 km/h, various slopes).

(b) Calisthenic exercises:

Exercise	Rate	Frequency
Deep Knee bends/squats	20/min	1 minute
1 minute rest.		
Touch Toes	25-30/min	" "
1 minute rest		
Twisting torso at waist so head and shoulders twist as far as possible 270° twist	25-30/min	1 min
1 min rest		
Raising arms from touching suit side seams to above the head repeatedly	25- 30/ min	1 min
1 min rest		
High kicks, approx 1 meter up	25/min	1 min
1 min rest		

(c) Assembling a slotted angle bench.

Heart rate and deep body temperature are continuously monitored throughout the test.

(5.) At the end of the test, the subject submits 10 urine samples for tritium assay over a 3-day period. The difference in tritium concentrations in urine between step 5 and step 1 give the tritium uptake which is expressed as ?Ci/L per MPC_a-h exposure.

(6.) The protection factor PF is calculated as:

$$PF= \frac{\text{Uptake without protection}}{\text{Uptake with protection}}$$

where "uptake without protection" is obtained by going through steps (1) through (5) with an essentially naked test subject. Measured protection factors for a range of protective equipment. Under field conditions, the protective factors are typically lower (as low as 100). A major reason for this is exposure incurred in a tritiated atmosphere while the worker is in transit to work locations and before air supply is connected to his plastic suit.